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AN INTERNATIONAL JOURNAL
FOR TECHNIQUES AND APPLICATIONS OF NUCLEAR SCIENCE

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THE ENERGY OF GAMMA RAYS

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Measurement of Nuclear Energy Change Values

An up-to-date review of studies of energy changes in nuclear reactions. Significance of these Q-values is explained in a discussion of characteristics of most widely used reactions. Actual experimental results, along with detection techniques, are included

By ERNEST POLLARD

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THE DISCOVERY that the bombardment of nuclei by energetic particles causes certain definite energy changes marks the beginning of the modern phase of nuclear physics. These definite values have been studied by a number of workers in the past quarter century and have given rise to many interesting discoveries, foremost among these being that of the neutron. A great many of these measurements have been summarized in Livingston and Bethe's excellent review article (*1*). However, as much work has been done since 1937, the date of the latter article, it is desirable to review the subject and bring it up to date.

Of great importance to the study of nuclear reactions is the fact that a fast, charged particle has a definite range in air (thus, 7 Mev alpha particles have a range of 6.9 cm in air or its equivalent in other absorbers). Basic work on this was done by Bragg. Of equal importance was the discovery that single, fast, charged particles cause scintillations in zinc sulfide crystals. The first use to which these two facts were put was the work on scattering by Rutherford and Geiger, and Marsden, which led to the idea of the nuclear atom. The next important use was the discovery by Rutherford of the transmutation of nitrogen in 1919.

Subsequent careful study of nuclear reactions by the observation of scintillations showed that the long-range particles responsible for scintillations are protons and that energy release is possible in the process. A further study was then made by Rutherford and Chadwick using apparatus illustrated schematically in Fig. 1. Alpha particles from the source—active deposit of radon [which is Ra(B + C)]—bombard a target placed at the end of an evacuated tube. The resulting transmutation protons pass through the thin foil sealing the vacuum and register scintillations on the screen. The energy of the protons can be measured by interposing foils between the target and the scintillation screen.

Results of this kind of study, and of cloud chamber observations by Blackett and Harkins, showed that the following

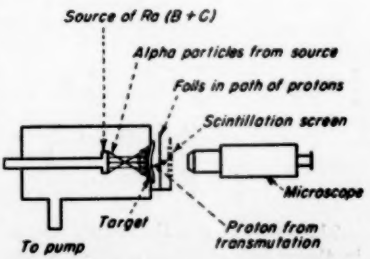

FIG. 1. Schematic of an early method of studying nuclear reactions. The protons are detected by scintillations and their energy measured by absorption foils placed in front of the screen

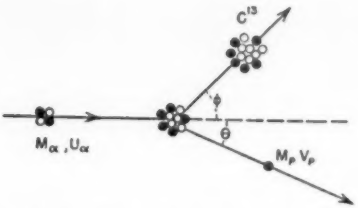

FIG. 2. Illustrating the process of a transmutation. The transmuting particle (here an alpha particle) is captured by the target nucleus (here B^{10}). The composite nucleus breaks apart into a final nucleus (C^{12}) at angle ϕ and a proton at angle θ

picture of the process is applicable: The alpha particle collides with and is *captured by* the nucleus it transmutes. The resulting composite nucleus has an exceedingly short lifetime, so short that it does not have time to travel the distance between two atoms in a solid. As a result, the composite nucleus either re-emits the alpha particle, which is then a *scattered* particle, or another particle, which indicates that a transmutation has taken place. In these early experiments, protons were detected as the new particle; later, neutrons were discovered in nearly identical experiments.

Because the lifetime of the composite nucleus is so short, energy and momentum are conserved for the whole process, which is illustrated in Fig. 2. Therefore, the following three equations hold.

$$\frac{1}{2}M_\alpha U_\alpha^2 + Q = \frac{1}{2}M_p V_p^2 + \frac{1}{2}M_n V_n^2$$

$$M_\alpha U_\alpha = M_p V_p \cos\theta + M_n V_n \cos\phi$$

$$0 = M_p V_p \sin\theta + M_n V_n \sin\phi$$

The quantity Q is the energy release in the reaction. It is positive if the mass of the product is less than the original mass. The value is given by $Q = c^2\Delta M$ where ΔM is the change of mass. The first equation then expresses the conservation of energy, the second and third that of momentum. By simply eliminating V_n and θ from these, there results

$$M_n Q = (M_n + M_p)E_p - (M_n - M_\alpha)E_\alpha - 2(M_n M_p E_\alpha E_p)^{1/2}\cos\theta \qquad (1)$$

Here M_n, M_p, M_α are the masses of the final nucleus, the proton, and the alpha particle, respectively; E_p and E_α the energy of the proton and alpha particle. As the equations were set up it would have been necessary to express Q in ergs; M_α, M_p, etc., in grams. However the final equation is of the same power in both mass and energy and therefore equation 1 is valid if all energies are in Mev and all masses are simple atomic masses. Usually the deviation from a whole number of the latter is ignored in working the equation, though not of course in using the relation $Q = c^2\Delta M$.

The next step forward was made by Bothe and Franz in 1929, after some preliminary work by Rutherford and Chadwick (*2*). Bothe and Franz showed that the bombardment of boron, which has two isotopes, led to the emission of *three* groups of protons, clearly separated from one another. While two of these could be the result of individual isotopes of boron, the third could not be so explained. Therefore the existence of at least two values of Q appropriate to one isotope must be supposed. This work was rapidly extended by Pose and by Chadwick, Constable and Pollard (*3*) who showed that alpha particle bombardment of a variety of light elements gave rise to multiple Q-values. It is the development and interpretation of this work which forms the subject of this review article.

Measurement of Nuclear Energy Changes

A considerable amount of work was done in the years 1930–1938 using alpha particle sources as a means of bombardment. The experimental procedure is

as follows: A target is bombarded by alpha particles of known energy and the groups of protons (or neutrons) are observed. Their energy is measured by absorption methods.

The following factors will then have to be considered. First, the use of equation 1 to calculate Q, the nuclear energy change, requires E_α to be definite. This in turn depends on the cleanliness of the source, the thickness of the target and the use of a small angular spread so as to diminish the variation of the range of the alpha particles in the target. Second, the energy of the proton must be definite which again requires a thin target and small angular spread. Finally, the angular condition must be good to permit $\cos\theta$ to be definite. This last is not quite as important as it seems because the term $2(M_n M_p E_\alpha E_p)^{1/2} \cos\theta$ is small compared to the others, except in the case of the very lightest nuclei.

These factors produce severe limitations because the yield of protons is quite small. It is well known today that most nuclear processes have an effective target area (cross section) which is about 10^{-24} sq cm (or one *barn*). Now consider a reaction with a cross section of one barn. The meaning of cross section is as follows. If a randomly distributed process takes place its occurrence can only be described as a chance, or probability. In terms of cross section, this is the ratio of the area of the bulls-eyes to the total target area and, if a target has a total area (as seen by the eye) of A and has a number of nuclei N in that area which can be hit, we have for the probability $N\sigma/A$ where σ is the cross section. Each alpha particle therefore has this chance of producing a proton. Now an alpha particle of energy 5 Mev travels 4.0 cm in nitrogen. The depth of the target is therefore 4.0 cm. The number N of nitrogen nuclei in an area A which can be hit is therefore the number in a volume $4.0 \times A$ cubic centimeters or $2.7 \times 10^{19} \times 4.0 \times A = 10.8 \times 10^{19} A$. The chance per alpha particle is therefore $\dfrac{10.8 \times 10^{19} A \times 10^{-24}}{A}$ which is about one per ten thousand.

These protons are emitted more or less at random so that by the time a reasonable angular spread has been achieved, corresponding to a reduction by one tenth of both source and emitted particles, the chance is one per million. Even so, the energy spread considered in E_α is from 0 to 5 Mev. If this is reduced to 0.5 Mev (still not very precise), the probability drops to one per ten million. Therefore, quite ordinary experiments, with crude geometry and poor energy definition require sources which emit more than 10^7 particles per second, *i.e.*, are in the millicurie region.

Such sources can be prepared, but not ones which are large enough to permit really thin targets and really small angular definition. Therefore most of this work from 1930–1938 must be classed as pioneer. The general results of this work can, however, be briefly summarized. Multiple Q-values were obtained for a number of reactions in which the largest element was between Be^9 and Ti^{48}. Notable exceptions are $Li(\alpha,p)Be$; $C(\alpha,p)N$; $O(\alpha,p)F$; and $A(\alpha,p)K$. The Q-values obtained were in general separated by between 1 and 2 Mev and were independent of the atomic weight. However, for the three reactions, $Mg(\alpha,p)$, $Si(\alpha,p)$, and $S(\alpha,p)$, Haxel showed that a "triad" of Q-values separated by less than 1 Mev exists.

Another interesting point is that increases of yield occurred for specific bombarding energies of the alpha particle. This was found to be the case in Be, B, N and particularly was shown to be so for Al by Chadwick and Constable (*4*) who found four such resonance levels for which the total yield of all groups was much higher.

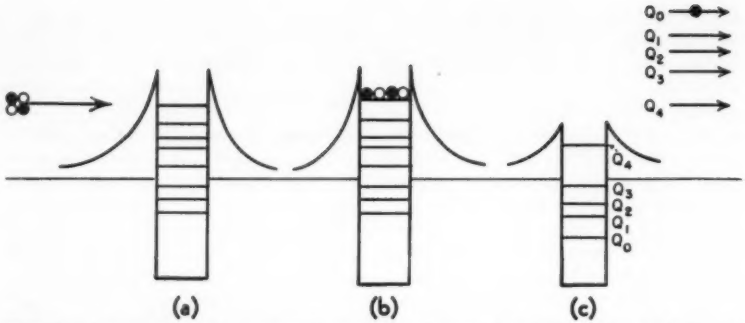

FIG. 3. A very schematic energy diagram for a transmutation of the (α,p) class. At (*a*) an alpha particle approaches a nucleus. The potential of the alpha particle in the target nuclear field is plotted and appears as a coulomb barrier plus a "well" of unknown character. At (*b*) the alpha particle is captured and is indicated as dissolved into its components (not necessarily true) but in a state of great excitation. At (*c*) the final nucleus is formed with a proton leaving. The proton leaves with maximum energy if the nucleus is in its ground state, Q_0, with less if the nucleus is excited, Q_1 to Q_4, the energy of the proton being directly determined by the nuclear energy levels. The singly charged coulomb barrier is less

A primary problem is the interpretation of the *Q*-values. The present-day opinion is that the smaller *Q*-values correspond to states of excitation in the *final nucleus* and that gamma radiation which is emitted should correspond exactly in energy and yield with the energies and yields of the corresponding proton groups. This is described in Fig. 3, where the initial nucleus with its potential barrier, the composite nucleus and the final nucleus in various states of excitation are shown. Very little direct evidence exists for this interpretation although such evidence is rapidly accumulating at present. One early experiment by von Baeyer (*5*) showed that coincidences were observed between gamma rays from boron, bombarded by alpha particles, and proton groups from the same element. This shows, at least, that there is a direct link between the proton groups and gamma radiation.

Work on the entire subject became more precise when artificially accelerated particles were used for the study of *Q*-values. This was begun by Cockcroft and Lewis (*6*) and has been continued by Oliphant, Kempton and Rutherford (*7*) and others at the Cavendish laboratory. Recently work has been done by Bonner's group at Rice (*8*), by Holloway and Moore (*9*) at Cornell and by several workers at Yale (*10*). These more precise studies will now be discussed.

Features of Various Reactions

Any accurately observed nuclear reaction will permit the determination of *Q*-values. The following have been most widely used: (α,p), (d,p), (α,n), (d,n), (d,α), (p,p). Each reaction has its own characteristics and these can now be considered. The basic points are *barrier penetration, energy release* and *range values.*

Barrier penetration is important for low-energy bombarding particles. It ceases to be important above 6 Mev for protons or deuterons or above 15 Mev for alpha particles. The repulsive coulomb barrier has a fairly definite height* for each element for a given

* The actual height depends somewhat on whether or not the nuclear reaction is produced

kind of particle. This height, which can be determined from the value of the nuclear radius and the nuclear charge, is $E_l = ZZ'e^2/r_a$ where Z is the nuclear charge, Z' the charge of the incident particle, e the electric charge and r_a the nuclear radius. Since $r_a = 1.5 \times A^{1/3} \times 10^{-13}$ cm where A is the nuclear mass, E_l can readily be found. Barriers range from less than 2 Mev for boron to 7 Mev for uranium for protons and double the latter figure for alpha particles.

A considerable degree of penetration occurs below the top of the barrier due to the "tunnel effect," a consequence of the wave nature of the incident particles. This is particularly true for protons, somewhat less for deuterons and decidedly less for alpha particles. In the case of alpha particles, their larger mass reduces this penetration. The penetration of the barrier can be seen in the excitation curves taken by Clarke and Irvine (*11*) and reproduced in Fig. 4.

Energy release. This is important because a highly negative Q-value will yield particles which have too little energy either to escape or be detected. Energy releases in (d,p) or (d,n) reactions are generally positive, (p,n) is commonly negative and (α,p) or (α,n) hover around zero.

Range values. A majority of the work done has involved the observation of proton groups in terms of range. If the range of the emitted particles is less than that of the scattered incident particles, then they are hard to detect, although new instrumentation may remedy this. This fact is closely linked with the energy release in the reaction but is also affected by the nature of the bombarding particle. Thus, a 10 Mev alpha particle has a range of 10.5 cm in air, whereas that of a 2.5 Mev proton is 10.4 cm. Thus, any proton of energy greater than 2.5 Mev could be observed. The situation is reversed for proton bombardment. Deuterons produce a positive energy release and have a relatively short range. Thus, for transmutation experiments, (α,p) and (d,p) are the easiest reactions to observe. In the case of lighter elements, where the low barriers permit very low bombarding energies, reactions which produce considerable energy release can be studied using proton bombardment even though alpha particles are emitted.

The long range of protons renders them very suitable for inelastic scattering experiments. This was first tried by Wilkins and Wrenshall and Kuerti and later by Dicke and Marshall (*12*).

Another important factor is that of contamination by high-yield elements. This is a serious matter, particularly where low-energy bombardment of elements of medium atomic number is attempted. The lighter elements, having low barriers, are much more prolific in yield than the heavier ele-

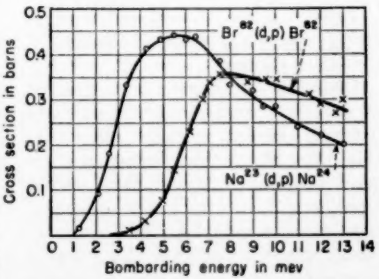

FIG. 4. Excitation curves for induced radioactivity by the $Na^{23}(d,p)Na^{24}$ and $Br^{81}(d,p)Br^{82}$ processes. The lower part of each curve corresponds to barrier penetration, the upper to transmutation after the top of the barrier has been reached

by particles colliding very nearly head on. This feature is described in terms of an impact parameter p such that $M_b U_b p < h/2\pi$ where M_b is the mass of the bombarding particle, U_b its velocity, and h is Planck's constant. p is the perpendicular from the target nucleus to the direction of the incident particle. For larger values of p, the barrier is higher by a term of the form $l\ (l+1)/r^2$ where $M_b U_b p = lh/2\pi$, r is the distance of the particle from the nucleus, and l is the orbital angular momentum quantum number.

TABLE 1

Summary of Some Reaction Processes

Reaction	*Energy Release for Highest Q*	*Remarks*
(αp)	up to +2Mev	Suitable because the alpha particle has low range, rather low yield
(α,n)	up to +2 Mev	Suitable if neutron detection is a properly managed technique
(d,p)	up to +6 Mev	High yield, suitable if deuteron energy is above 3 Mev
(d,n)	up to +5 Mev	High yield, suitable if neutron detection is available
(d,α)	up to 15 Mev	Suitable if deuteron energy is below 3 Mev otherwise the alpha particles are masked
(p,α)	up to 10 Mev	Only in the lightest elements
(p,p)	zero	Very suitable at proton energies above 5 Mev

ments and a one percent impurity may give a yield equal to that from the legitimate target. Carbon, oxygen and nitrogen are especially dangerous impurities in this respect.

Table 1 gives a broad summary of several reaction processes in the light of the above remarks.

Detection technique. The method of detection must clearly differentiate between charged particles and radiation. For this reason, Geiger counters are not suitable. Most of these studies have been done with either an ionization chamber and high-gain amplifier which directly amplifies the primary ionization, or a proportional counter which uses a single "avalanche" of ionization-by-collision to multiply the charge collected. In the last few years, the proportional counter has gained in popularity. Its chief advantage is the absence of a temperamental, microphonic amplifier on the one hand, and rapidity of counting action on the other. The use of such counting has been described in many articles. Reference is made to papers by Benson and Martin (*10*) as giving the most recent methods.

It is very often necessary to reduce unwanted background counts. For example, in a cyclotron running on deuterons, there is always a large yield of neutrons from the $D(d,n)He^3$ reaction, among others. These bear no relation to the reaction being studied and hinder the measurements. If a single counter is used, with a target near the cyclotron "can," the density of counts due to these neutrons may well mask a weak, well-collimated proton group. By setting two counters in line with the target and observing double coincidences it is possible to reduce nearly to zero the background caused by neutron recoils. In this way, it was possible to observe protons from the rare isotope C^{13} in spite of a considerable neutron background.

The detection of neutrons is easy, but not the measurement of their energy. This has to be done either by a threshold method, as in the (p,n) reactions, where the proton energy which just produces neutrons is observed, or by using recoil protons or helium ions. For this purpose, cloud chamber or photographic methods have been the most popular because the process of using recoil requires that only those recoil nuclei which are in a known direction be measured and a technique which enables the process of recoil to be seen is very desirable. Cloud chamber work has been done by Bonner and his group (*8*) and the photographic method has been used Richards (*14*) and by Peck (*15*). The difficulty in photographic detection is the calibration of the emulsion which has to be done by observing the tracks of known-energy protons in

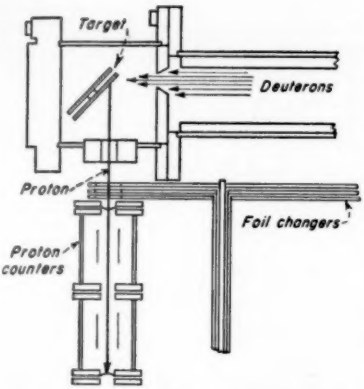

FIG. 5. Arrangement of beam, target and counters for the observation of nuclear reactions. The foil change permits the observation of the number of protons passing a given absorption, which in turn gives the energy of the proton group responsible

the emulsion: the advantage is that data can be taken simultaneously at several angles and, once taken, is permanently recorded. At the present time the great majority of the observations of Q-values has been taken using (α,p), (d,p), and (p,p) reactions.

Experimental Results

(d,p) reactions. A typical bombardment arrangement is shown in Fig. 5 which is taken from a paper by Martin. This is set up for what is called 90 deg bombardment. The beam enters through a defining aperture and is incident on the target. The protons from the target emerge through a hole sealed with an aluminum foil, go through a variable absorption determined by a foil changer, and into the two counters of a double coincidence arrangement.

The target must be designed with great care. The most important factors to be considered are thickness (or rather thinness), support, stability to heat, and electrical conductivity. The target must not have so great a thickness that its absorption of the beam causes a spread in the bombarding energy and so impairs resolution. A favorite method of target preparation is evaporation in vacuum. The target support must not be transmuted itself, or the interpretation of results becomes difficult. This is one reason why high-energy cyclotrons are not necessarily an asset since all elements are roughly equally transmuted by a beam of 10 Mev deuterons. To overcome this, a careful definition of the beam and the direction of observation can be employed. Then the support does not get bombarded or, if it does, the counter cannot see the emerging protons. In this way, a thin foil can be suspended on a frame and yet be bombarded satisfactorily.

Stability to heat is necessary unless very weak beams are used. Since high yields are generally observed in (d,p) reactions, the beam used can be less than one tenth microampere. Even so up to a watt can be dissipated in the target which, if it is thin, may evaporate unless it is adequately cooled. A thin, transparent target with very little absorption intercepts much less energy and can very often be used with quite intense beams.

Finally, the beam hitting the target must be measured; therefore, the target must conduct. If it does not, some other method of beam measurement has to be used. One method is to monitor the beam by counting protons at a fixed absorption, or, if gamma rays are emitted, counting the gamma rays produced by a second counter.

The procedure used depends on whether the maximum proton energy in a reaction is to be measured or whether the group structure of the protons is to be analyzed. For the former, the greatest possible yield with the least background is needed. For the latter, the best possible energy resolution is essential. It is usual to

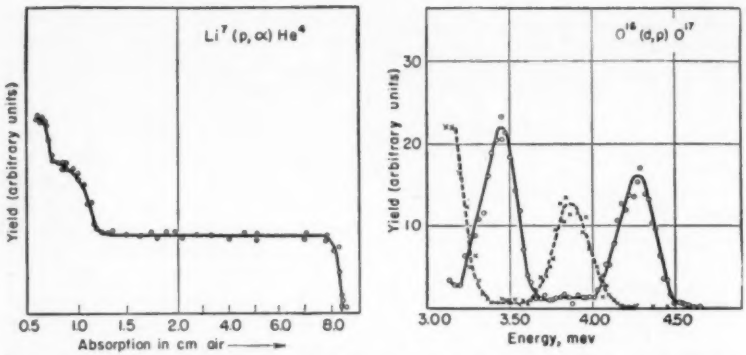

FIG. 6. (left) A yield versus absorption curve for low recording bias. Reaction $Li^7(p,\alpha)He^4$, data due to Oliphant, Kinsey and Rutherford; (right) a curve for the reaction $O^{16}(d,p)O^{17}$ with high recording bias giving a peaked curve. Data due to Pollard and Davison. The dotted curve is for a lower bombarding energy and shows the variation of relative yield of the two groups

secure one or other of these conditions by varying the recorder bias. A word on recorder-bias action is thus in order.

There is a definite relation between the range and the energy of a charged particle. This is fully discussed in Livingston and Bethe's review article (*1*) and is the basis for energy measurement. A charged particle produces ionization at the rate of 33 electron volts per ion pair. Therefore, the ionization produced in a counter of definite length at a known pressure is simply the energy lost in the counter in electron volts divided by 33. For a counter of length 2 cm having a pressure of 15 cm argon, the equivalent absorption is $\frac{15}{76} \times 2 \times$ argon equivalent and is very nearly 0.4 cm air because argon is very nearly equivalent to air in absorbing particles. Consulting range-energy relations, it can be seen that protons having a residual range of 0.4 cm on entering the counting volume lose 260,000 ev, protons of residual range 5 cm lose 90,000 ev, and protons of residual range 200 cm lose 40,000. A proton which just completes its path in the counter therefore produces nearly three times the ionization of a proton 5 centimeters from the end of its range and six times that of a 200 cm proton. Since the counter is proportional, this effect appears in the output and, by biasing the recording circuit, the record can be confined to protons very close to the end of the range. Because circuits which are well-regulated can be made very precise, it is easily possible to bias the recorder to register within one centimeter of the end of the proton range. This process, known as "peaking" was originated by Chadwick and Constable and has been widely used since. By biasing the recorder to pass small voltage pulses, it is possible to count all the protons, unless the range is very great. In the latter case, the problem of distinguishing a fast proton from a superposition of several slow recoil electrons becomes serious. This starts to be troublesome in the case of 100-cm protons and requires the design of fast-acting counters which avoid the piling up of several discharge processes and give abnormally large output voltages.

Examples of results using the two methods of counting are shown in Fig. 6.

The first curve, taken by Oliphant, Kinsey and Rutherford (*7*) shows the definite group of alpha particles from the bombardment of Li^7 by protons; the second, taken by Davison, (*10*) shows the two groups of protons from $O^{16}(d,p)O^{17}$.

Once the absorption curve is plotted, the group structure is generally apparent. Peaked absorption curves taken for Ne(*d*,*p*)Ne by Elder, Motz and Davison (*10*) are shown in Fig. 7. To calculate *Q*-values accurately, a certain amount of analysis of the data is needed. Tolerably good values can be obtained quickly by measuring the extrapolated numbers range of each group. This is then converted into energy by consulting the range-energy curve, and the *Q*-value obtained from equation 1. In principle, this is all that is necessary. In practice several factors have to be determined. The first is the spread in energy of the beam. This can be measured rather roughly by range measurements and more accurately by magnetic analysis. The energy spread of a 3.8 Mev cyclotron deuteron beam without magnetic analysis is about 200 kev. The second factor is target thickness; the third, geometrical imperfection due to finite solid angle. Finally there is a theoretical straggling of each group due to collision fluctuations in passing through the absorbers. A procedure for correcting for these factors has been devised by Livingston and Bethe. A further careful discussion has been given by Holloway and Moore (*9*). The principal problem is that of determining how these various factors affect the range measurement, taking into account the fact that the counter bias is set to produce a peaked absorption curve. To make the corrections fully it is necessary to plot a non-peaked (or integral) curve and from this deduce the correction to the range for the peaked curve. This is laborious, but straightforward.

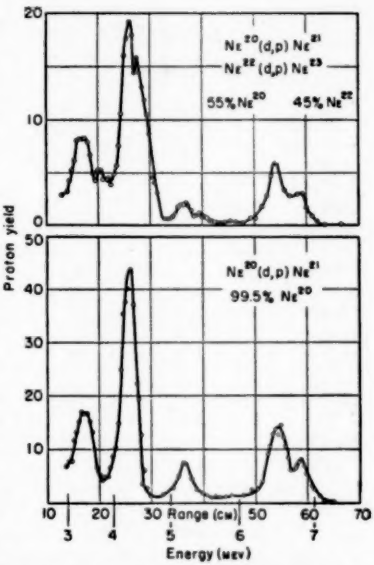

FIG. 7. Peaked absorption curves for protons from two mixtures of neon isotopes bombarded by deuterons. The group structure is clearly visible both for $Ne^{20}(d,p)Ne^{21}$ and $Ne^{22}(d,p)Ne^{23}$. Data due to Elder, Motz and Davison

A summary of reasonably careful work on (*d*,*p*) *Q*-values is given in Table 2. A great many of the elements are complicated by the presence of several isotopes which render interpretation difficult. The recent increase in the availability of separated stable isotopes will greatly improve the certainty of assignment of groups and increase the scope of the table. Some separated isotope work is included in Table 2, particularly for Li, C, and Ne. A partial separation was used for chlorine which enabled some assignment of groups to be made. Some of the *Q*-values of $Cl^{35}(d,p)Cl^{36}$ are certainly masked by those from $Cl^{37}(d,p)Cl^{38}$.

One or two elements show a group structure which has not yet been resolved. This is the case for $Al^{27}(d,p)Al^{28}$ and $P^{31}(d,p)P^{32}$. The former has been

TABLE 2

(d,p) Q-values

Reaction	*Q*	*Reference*
H^2H^3	3.98	17
Li^6Li^7	5.02, 4.57	18
Be^9Be^{10}	4.59, 1.11	19
$B^{10}B^{11}$	9.14, 7.00, 4.71, 2.39	6, 10*a*
$C^{12}C^{13}$	2.38	6
$C^{13}C^{14}$	5.82, 0.58	8*b*, 10*d*
$N^{14}N^{15}$	8.55, 3.11	9
$O^{16}O^{17}$	1.95, 1.12	6, 10*g*
$F^{19}F^{20}$	4.3, 3.6, 3.3, 2.95, 2.4	20
$Ne^{20}Ne^{21}$	4.48, 4.17, 2.73, 1.65, 0.90	10*c*,*h*
$Ne^{22}Ne^{23}$	2.89, 1.90, 1.23	10*h*
$Na^{23}Na^{24}$	4.76, 4.38, 3.50, 1.38	
$S^{32}S^{33}$	6.50, 5.74, 4.62, 4.28, 3.54, 3.25, 2.75, 2.14, 1.74, 1.23, 0.78, 0.12	22
$Cl^{35}Cl^{36}$	6.31, 5.35, [some missing], 1.50	23
$Cl^{37}Cl^{38}$	4.02, 3.02, 2.10	23
$A^{40}A^{41}$	3.83, 3.19, 2.55, 1.90, 1.53, 1.01, 0.54, 0.04	24
$Ca^{40}Ca^{41}$	6.30, 4.51	25
$Sc^{45}Sc^{46}$	6.78, 4.48	26
$V^{51}V^{52}$	7.80, 5.33, 3.10	26
$Mn^{55}Mn^{56}$	4.76, 3.69, 2.99, 1.15, 0.38	10*e*

studied by Schultz, Davidson and Ott (*10*), the latter in unpublished work by the author. It is expected that with magnetic analysis of a deuteron beam the resolution can be greatly improved and a definite group structure found. In any event the separation of *Q*-values is not much greater than 0.3 Mev and probably less.

(α,*p*) reactions. The great majority of (α,*p*) work has been done with natural alpha particle sources with rather poor resolution. There seems, however, to be a rather greater spacing of *Q*-values in many cases than is found for (*d*,*p*) reactions with the result that fairly definite group structure has been established. Very recently, some of these reactions have been reviewed using doubly charged helium ions accelerated in a cyclotron. These can be obtained in better yield than natural alpha particles and are collimated. Such beams are not more homogeneous in energy unless magnetically analyzed.

The recent data tend to confirm the earlier findings, with some added group structure being apparent. Since there is a very complete account of the early (α,*p*) work in Livingston and Bethe no detailed account will here be given. Some recent work is tabulated in Table 3. The general feature of (α,*p*) work is a *Q*-value difference which lies between 1 Mev and 2.3 Mev. The most definite regularities are found between odd and even product nuclei as evidenced by the reactions $F^{19}Ne^{22}$, $Na^{23}Mg^{26}$, $Al^{27}Si^{30}$, $P^{31}S^{34}$, $Cl^{35}A^{38}$ versus $Mg^{24}Al^{27}$, $Si^{28}P^{31}$, $S^{32}Cl^{35}$. The three last show small *Q*-value spacing (less than 1 Mev) while the other five show an average spacing of 1.45 Mev. The most carefully studied single reaction has been $Al^{27}(\alpha,p)Si^{30}$ which has set the pattern for the work on other elements.

(*d*,*n*) and (α,*n*) reactions. Both these reactions are shadowed by the difficulty of measurement of neutron energy. Satisfactory early work was done by cloud chamber detection of hydrogen recoil nuclei produced by neutron impact. The work is very laborious and involves many expansions and much analysis of the data. This analysis is required for two major reasons. The first is the energetic relationship between the deuteron and the recoil proton which depends on the angle at which the collision is made. It is therefore necessary to be sure that neutrons can only enter the cloud chamber in one definite direction—from the target (*i.e.*, no scattered neutrons

TABLE 3

(α,p) Q-values

Reaction	*Q*	*Reference*
$Al^{27}Si^{30}$	2.22, −0.06, −1.44, −2.50	10*f*, 27
$Sc^{45}Ti^{48}$	−0.3, −1.4, −2.6	28
$Ti^{48}V^{51}$	1.10, 0.00, −3.63	29

should be present)—and then only recoil protons along, or nearly along, that direction are measured. The second reason affects the yield and is caused by the variation in collision cross section between a neutron and a proton as the neutron energy varies. The cross section gets less with energy which therefore gives less weight to the high energy tracks. In addition the finite thickness of the emulsion diminishes the chance of observing a high energy track.

The cloud chamber has one serious disadvantage in that most versions have an air equivalent not exceeding 30 cm. This means that a complete survey of neutron group structure requires a specially constructed high pressure chamber or else a system of foils across the expansion region.

Serious application of photographic detection was first made by Wilkins (*12*). The rather formidable technique of development and observation, plus the need for special emulsions, rendered the method unpopular until its further exploitation by Powell (*16*) in England and Richards (*14*) at Rice Institute. Today two suitable emulsions are available, the Ilford and the Eastman proton plates. The former is thicker, which makes it easier to use where long tracks are involved.

TABLE 4

(d,n) and (α,n) Reactions

Reaction	*Q*	*Reference*
$Be^{9}(d,n)B^{10}$	4.2, 3.7, 2.2, 0.9	8*a*
$C^{13}(d,n)N^{14}$	5.5, 0.4	8*b*
$B^{11}(d,n)C^{12}$	13.4, 9.0	8*a*
$F^{19}(d,n)Ne^{20}$	10.8, 9.33, 6.62, 5.39, 3.53, 1.84	8*c*
$Al^{27}(\alpha,n)P^{30}$	−2.93, −3.91	30

The application of this method to (d,n) and (α,n) reactions is now being made. The process is laborious because a complete group survey needs the measurement of between one and ten thousand tracks. These have to be measured individually and it is hard to maintain a consistent rate of measurement in excess of a hundred per day. Therefore a single "run" takes about a month to count and if the conclusions show inconsistencies, which mean that a repetition of the data is desirable, it may take two more months to correct. Nevertheless some very good work is now being done, with definite and reproducible results. The expansion of effort in this field is very rapid and in a year or two Table 4, for (d,n) and (α,n) reactions, will be much longer.

(p,α) and (d,α) reactions. The fact that the alpha particle has very little mass excess compared to protons and deuterons means that both (p,α) and (d,α) reactions have positive Q-values. This is especially true for (d,α) reactions. On the other hand the alpha particles have, in general, less range than protons or deuterons and so require special detection technique. However, for many of the lighter elements, there exist nuclei of abnormally high mass and by use of these as target elements very large Q-values are observed. The groups of alpha particles then have a long range and are easily detected, particularly if the bombarding proton or deuteron is of low energy and hence short range. As the barrier gets higher, more energetic bombarding particles are needed and their range rapidly gets greater. For this reason, compara-

TABLE 5

(d,α) Reactions

Reaction	*Q*	*Reference*
$F^{19}O^{17}$	10.22, 9.39, 7.17, 6.23	30
$Na^{23}Ne^{21}$	6.75, 5.15	20
$Cl^{35}S^{33}$	9.1	22

tively little observation of (d,α) processes has taken place above fluorine.

A second reason for expecting alpha particle emission to be less prominent as the nuclear charge increases is the coulomb barrier. The alpha particle has to escape over or through this barrier which is twice as high as for a proton. Therefore competing (d,p), (d,n), (p,p) or (p,n) processes would be expected to be favored for elements above, say Ca^{40}. This does not mean that the reactions do not occur, but presents a considerable detection problem. Table 5 shows some results of (d,α) reactions.

(p,p) reactions. This term is employed to denote inelastic scattering of protons. It is correctly employed because the process involves the capture of a bombarding particle, the formation of a composite nucleus and the emission of a particle, leaving the final nucleus

TABLE 6

(p,p) Reactions

Reaction	*Q**	*Reference*
Be^{9}	−2.42	31
Al^{27}	−0.91, −1.90, −2.92, −3.34	12*d*
Mg^{24}	−1.24, −2.56, −3.59	12*d*
S^{32}	−1.86, −3.63	12*d*
Cr^{52}	−1.28, −2.74, −4.07	12*d*

* Zero in all cases in addition to the values quoted

either in the ground state or a state of excitation which reverts to ground by the emission of gamma radiation. There is no reason why (α,α) or (d,d) reactions should not take place, and in fact one definite (α,α) process is known, $Li^{7}(\alpha,\alpha)Li^{7}$. Most of the work has been done with protons for two reasons. The first is the ability of a low energy proton to emerge through the low, single charge coulomb barrier: the second is the fact that protons have long range so that even scattered protons which have lost energy, still travel further than reaction products such as alpha particles.

One major problem of such research is the problem of obtaining the scatterer in a thin foil form. If a thick foil is used the energy of the protons which pass in and are scattered out varies very greatly, purely on account of absorption in the foil. Therefore a thin foil has to be obtained which must in turn be supported by a frame which is not bombarded by the beam. For such elements as aluminum this is no problem since they can readily be rolled into foils. But for targets of chlorine or fluorine, the problem is more difficult.

It is advantageous to scatter through the foil, rather than in and out as in a reflection process. This gives less total absorption in the combined incident and scattered path.

In Fig. 8 an absorption curve for scattered protons, taken by Dicke and Marshall, is shown. The elastically scattered protons naturally have a *Q*-value of zero. The inelastically scattered have negative *Q*-values. These correspond to states of excitation in the target nucleus.

Table 6 shows the results of inelastic scattering work.

Some other reaction types. Before concluding this summary of experimental methods of measuring *Q*-values, some other reaction types can be considered. Reactions in which the inci-

dent particle is a neutron are of much interest but are extremely hard to study. Only photographic or cloud chamber technique is available for direct study and the work is complicated by the large amount of normal recoil nuclei present plus the danger of hydrogen contaminant. Any cloud chamber, or emulsion, contains some hydrogen which accordingly gives recoils. These have to be recognized and rejected, which is a complex process, certainly until much more experience in deuteron reactions is available. Some indirect methods of studying these reactions have been devised. Thus Wilhelmy (*32*) has bombarded gases enclosed in an ionization chamber attached to a linear amplifier and measured the sizes of deflections. These fall into many small groups which indicate the operation of some quantum process. The interpretation is difficult because such work has been done with sources of neutrons of rather mixed energy and it has been hard to say whether the observed group structures is due to the primary neutrons, to selective neutron absorption or to the production of a nuclear reaction with many Q-values and correspondingly many excited states of the product nucleus. One such element which has been studied is nitrogen for which two reactions can occur, $N^{14}(n,p)C^{14}$ and $N^{14}(n,\alpha)B^{11}$. The distribution of amplifier responses when neutrons from RnBe and PoBe sources are allowed to pass through the gas is shown in Fig. 9.

This technique could well be applied using homogeneous neutron sources such as photo neutrons from Na^{24} and deuterium. No published work is available on this.

The use of He^3 as a bombarding particle has much promise. Its range is short, yet it carries great mass energy and should give very large Q-values. The observation of protons corresponding to very high states of excitation of

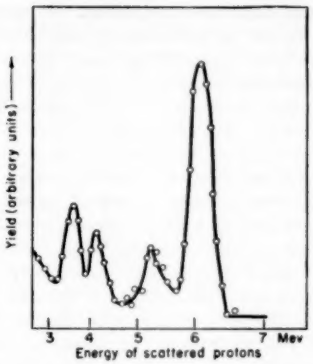

FIG. 8. Inelastic scattering of protons from aluminum. The elastically scattered protons have the greatest range while the inelastically scattered protons fall into groups having negative Q-values corresponding to excited states in Al^{27}. The data are due to Dicke and Marshall

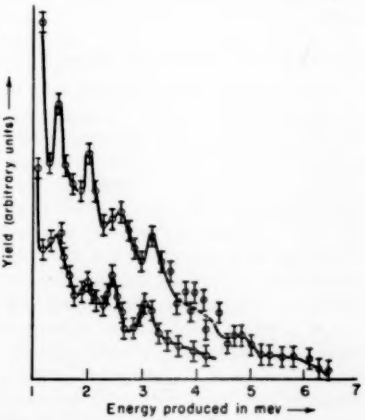

FIG. 9. Distribution of amplifier responses when neutrons from Rn + Be (upper) and Po + Be (lower) produce reactions in a nitrogen-filled ionization chamber. These are in groups indicating the operation of nuclear energy levels of some form, partly to two reaction processes $N^{14}(n,\alpha)B^{11}$ and $N^{14}(n,p)C^{14}$, and partly to resonance as the charged particle leaves the composite nucleus. Data due to Wilhelmy

the final nucleus should be very easy.

This is one field of nuclear physics in which high bombarding energy is not necessarily an asset. High-energy bombardment is known to produce complex reactions, known among cosmic ray workers (who are quite used to high energies) as *stars*. These stars may or may not include neutrons and the experimental analysis therefore must involve great maturity of knowledge to enable the actual process taking place to be diagnosed. When this is done it is doubtful whether the results will be of interest by reason of the measurement of nuclear energy levels but rather in terms of the probability of nuclear disruption in differing ways. This is of course of high importance, but can only concern us here insofar as it affects the discovery of a definite nuclear theory. A great deal of low energy (0–5 Mev) work remains to be done and therefore high energy work will lag.

Technical Improvements Needed

The work here described can be said to be primitive nuclear spectroscopy. It shows many of the properties of that science. Thus characteristic nuclear spectra are already noticeable, particularly that of nitrogen, with a large Q-value followed by a long interval before the first excited state. This has been used in several experiments to check on the purity of a target and could, in principle be used as a method of analysis. It could readily be applied to determine the proportion of two isotopes.

However, while characteristic nuclear spectra are already available, the theoretical requirements of a science of spectroscopy are not yet fulfilled. This is in part due to the fact that theory has not come to the aid of interpretation of Q-values and in part due to the lack of experimental precision in taking data. Thus, while the Balmer series has been of the utmost use in the interpretation of the atom, the interpretation would have been much harder if only a few of the lines were clearly resolved. Therefore better energy resolution is of great importance. This is attainable by magnetic analysis as regards beam energy, but is difficult of attainment as regards target thickness. Very careful work is needed to establish that the Q-values found are actually all the Q-values present in the reaction.

Direct Observation of Gamma Radiation

To each of the excited states of the product nucleus, there must correspond gamma radiation produced when that nucleus reverts to a state of lesser excitation. The direct observation of such radiation is therefore of great importance because it confirms the interpretation of the energy level schemes deduced from the measurement of Q-values.

The presence of such gamma radiation has been shown by many workers. Crude energy measurements by absorption have shown that the range of energy values found for gamma radiation is about right. Among the lighter elements several definite gamma-ray values have been linked clearly with excited states known to be present in a nucleus. Thus the deuteron bombardment of Li^7 gives two groups of protons separated in energy by 455 kev. It has been shown that a gamma ray of this energy is produced in this reaction and also by the two processes $Li^7(p,p)Li^7$, $Li^7(\alpha,\alpha)Li^7$. This state of excitation is therefore fully recognized. On the other hand other gamma rays have been observed which are not yet assigned to excited states as determined by the observation of particles.

A start on the correlation of gamma rays and Q-values has been made by Alburger (*34*). The gamma rays emitted in the six (α,p) reactions from Na to S have been observed and their

energies measured, first, by absorption in lead and, second, by observation of the range of Compton recoils in aluminum. The latter method gives ten percent accuracy and has enabled the correlation of several Q-value intervals with the measured gamma rays.

One difficulty of this kind of work is the fact that in a nuclear bombardment there are several simultaneously occurring reactions and it is hard to assign the observed gamma radiation to the correct process. This difficulty can be overcome by increased precision which should show clearly which gamma rays are appropriate to certain Q-values, and by coincidence counting. A word on this last is in place.

Coincidence Counting of Protons and Gamma Rays

If a group of protons corresponds to an excited state of the final nucleus, there should be a time coincidence between any proton of the group and a gamma ray. This will not be exact, for there is a finite lifetime to the excited nucleus; however, the expected time is of the order of 10^{-12} sec so that the coincidence is pretty close. If such coincidences are observed, it is excellent evidence that the assignment of groups is correct, and if, moreover, the gamma-ray energy can be measured for coincident gamma rays it should be possible to make a complete assignment of gamma rays to proton groups. Such a study is very attractive: it proves in practice to be hard though work has been done by von Baeyer (*5*), Benson (*10*) and Davison.

The difficulty arises from solid angle considerations. If a true coincidence occurs, it is likely that any predictable angular correlation can be observed. Therefore the number of observed coincidences is less than the actual by the product of the solid angles for detection of proton and gamma ray. If, in addition, the efficiency of a gamma ray counter is below 2% there is an additional loss of a factor of 50. The end result is that only about one in ten thousand true coincidences can be counted. This renders counting slow and tedious. In addition, if the resolving time, T, of the counters is long the number of random coincidences, $2TN_pN_\gamma$ (where N_p and N_γ are the counting rates for proton and gamma ray, per second) can exceed the number of true coincidences by a large factor. This again requires either T small, or N_p, N_γ small and means slow counting or good resolution.

The recent development of high-speed counting methods has greatly accelerated the process of coincidence counting and it is likely that the coming year will see a good deal of this kind of work.

Variation of the Yield of Groups

The relative yield of particles in the various groups changes with (*a*) kind of reaction, (*b*) bombarding energy, and (*c*) angle of observation. The first and second are shown clearly in work on the ground and first excited state of O^{17}. This nucleus can be formed by the three reactions $O^{16}(d,p)O^{17}$, $N^{14}(\alpha,p)O^{17}$, $F^{19}(d,\alpha)O^{17}$. By all three processes, Q-values are observed which agree with a first excitation energy of 0.85 Mev. However, the first and last processes give nearly equal yields for the two groups whereas the (α,p) process gives very much less for the first excited state. All three are for 90 deg observation. The first reaction shows a steady decrease in the yield of the first excited state as the bombarding energy of the deuterons increases from 1 Mev to 6 Mev. In general the groups of higher excitation are more populated, though this rule has many exceptions.

Evidence for the variation with angle of observation has recently been found by Heydenburg and Inglis (*35*) and Davison (*22*) who find the group

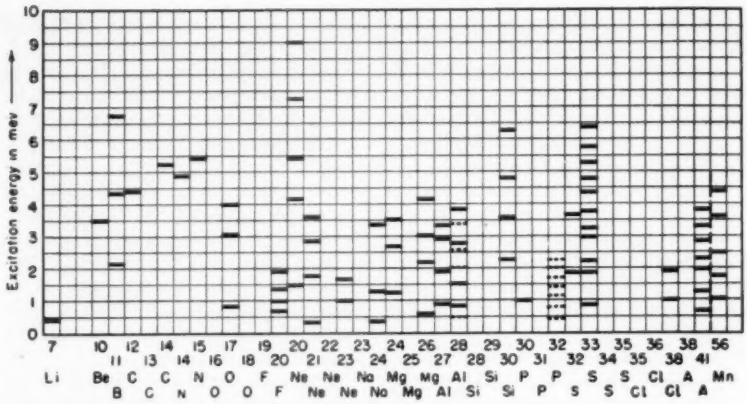

FIG. 10. Chart showing some of the known states of excitation of nuclei. Not all known states are given, but the majority of the information obtained from accelerated particle bombardment is shown. The dotted levels for Al^{28} and P^{32} indicate unresolved, closely spaced levels. No definite behavior can be seen except that the completion of a neutron shell at C^{14} and N^{15} seems to require very large spacing

structure yield from $S^{32}(d,p)S^{33}$ to change greatly when the observation is changed from 90 deg to 0 deg.

General Character of Q-values

A chart showing a number of energy levels deduced from Q-values is shown in Fig. 10. There is no particular pattern noticeable at first glance. Perhaps the most striking feature is the regularity of spacing; the spacing varies from element to element. Some odd-even variation is visible.

From the highest Q-values, mass differences can be deduced providing sufficient reactions are available. Satisfactory tabulation of masses up to Ne has been achieved but sufficient post-war work to ensure that true values are given has not been completed beyond Ne. This is an important aspect of research in nuclear energy changes.

More careful inspection shows that there is some evidence for shell structure in nuclei. Thus where a shell is completed by the reaction process there appears to be wide spacing, as for N^{15} and C^{14}, where the P-shell is completed for neutrons.

No theoretical interpretation of the levels is at present available. This is not altogether surprising because the particles comprising the nucleus are still under debate. It is to be hoped that a theory will be forthcoming which will be able to give a convincing account of nuclear structure.

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"I have heard and read numerous statements that many corporations buy valuable inventions to suppress them, but no one cites specific cases. I myself do not know of a single case."

—Thomas A. Edison

"The myth that patents on valuable inventions are bought up and suppressed to prevent competition will probably live as long as human nature remains what it is.

"Yet there is very little, if any, truth in the myth. The practice, if not absolutely non-existent, is so rare as to be entirely negligible. Rigid legal proof of this may be impossible since it involves proof of a negative character over a field so vast that no individual could speak with certainty about every procedure in it. However, the weight of evidence overwhelmingly supports the statement."

—Frank B. Jewett, The Journal of Commerce, *March 11, 1948*

ENERGY LEVELS OF LIGHT NUCLEI

This paper is based on two review articles* from the Kellogg Laboratory, appearing in the January-April, 1948, issue of the *Reviews of Modern Physics*, which is dedicated to Dr. R. A. Millikan on the occasion of his 80th birthday. Although the material presented represents the work of many laboratories, the field covered is that which has been of particular interest to the Kellogg Laboratory group and to which they have made some contribution. We feel that this article is particularly appropriate to the present celebration because, since the very inception of the laboratory, the work has benefited by the whole-hearted support and enthusiastic interest of Dr. Millikan. It has been a great pleasure to all the members of the group to have had the opportunity to join in the celebration and to record their indebtedness to him.

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One of the fundamental problems of nuclear physics concerns the study of the various states of atomic nuclei. It has been known for many years that the nucleus can exist for appreciable times in quasi-stationary states with energies considerably in excess of that possessed in the ground state, or normal configuration. Some of the general features of such excited states and of the transitions from one to another and to the ground state admit of at least qualitative understanding in terms of the interactions of the elementary particles comprising the nucleus. As more complete and more reliable experimental data become available, one can hope eventually to obtain information of quite fundamental character regarding the structure of the nucleus, just as the study of atomic spectroscopy contributed to an understanding of the structure of the atom. In the second article referred to above, we have attempted to collate the currently available information on the levels of certain of the light nuclei; it is the purpose of the present paper to outline the principal methods by which the data have been gathered and to summarize some of the results which have been obtained.†

In the light elements—those with atomic number up to eight or ten—the lowest excited states presently known lie from 0.4 to five or six million electron volts above the ground states. In general, the spacing between states decreases with increasing degree of excitation and at energies of ten to twenty Mev they may merge into what is essentially a continuum of overlapping states. The region of interest, then, lies between a few hundred thousand and about twenty million volts of excitation, and the problem of nuclear spectroscopy consists in obtaining excitations of this order and of observing the characteristics of the states so excited.

* Gamma Radiation from Excited States of Light Nuclei, by W. A. Fowler, C. C. Lauritsen and T. Lauritsen; Energy Levels of Light Nuclei, by W. F. Hornyak and T. Lauritsen.

† Specific references to original publications are given in the *Reviews of Modern Physics* articles.

The processes involved in nuclear reactions have some similarities to those of atomic spectroscopy and several important differences. In the study of atomic states by optical methods, one ordinarily excites the system by the addition of an amount of energy—of the order of a few electron volts—in the form of thermal energy transmitted through molecular collisions, kinetic energy by electron or ion bombardment, or quantum energy by irradiation with light of appropriate wavelength. As long as the excitation is less than the first ionization potential, the atom is unchanged except that the configuration of the electrons has been modified in such a way that they no longer are in the condition of minimum energy. The decay of this excited system proceeds with emission of radiation with wavelengths characteristic of the transitions between various possible states made in the course of the return to the minimum energy arrangement.

Excited states of atoms can also be observed in the phenomenon of resonance absorption. If an atom is irradiated with white light, certain wavelengths will be selectively absorbed. These absorption lines can be associated with the energy differences between the various states and are used to map the level system. In the case either of emission or absorption spectra, the characteristic feature is the regular variation of the intervals between successive lines of a given series, which, of course, is a consequence of the regularity of the spacing of the stationary states of the atom and is due to the existence of the central field. Another feature of importance is the existence of "series limits" which represent the energy of transitions to the several stationary states from a configuration in which one electron is completely removed from the atom. Beyond this excitation—the ionization potential—no discrete states exist: any value of the energy is permitted.*

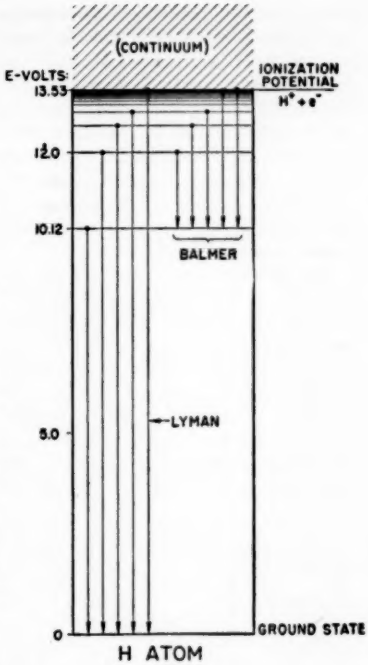

FIG. 1

In Fig. 1, some of these features are schematically presented for a simplified atom. The stationary states are represented by horizontal lines, on a vertical energy scale, in electron volts of excitation. The lowest level corresponds to the energy of the normal hydrogen atom, in which the total energy of the atom is 13.53 electron volts less than that of the ion. Transitions from excited states to the ground level result in the radiation of the spectral series known as the Lyman lines; the series limit, *i.e.*, the shortest wavelength line,

* The cases in which two valence electrons or inner electrons are excited are exceptions: in these cases states of the system exist above the minimum ionization potential; in fact this phenomenon is in many respects analogous to the behavior of the nucleus.

is equal to the wavelength corresponding to the ionization potential of 13.53 ev, or 915 A.U. Other series arise from transitions to higher states and their limits correspond to the differences between the ionization potential and the end states. If the atom is excited beyond 13.53 ev, the electron is thrown off, and the system may be thought of as having positive energy with any value being permitted.

The situation in a nucleus can be represented diagrammatically in much the same fashion (Fig. 2). Here we can speak, first of all, of the lowest, or ground state, which is stable for most of the nuclei found in nature but which, in the case of radioactive nuclei, can be unstable with relatively slow α or β-decay (β^+, β^-, K-capture). (In rare cases as He^5 and Li^5, the ground state decays by neutron and proton emission.) We can also speak of several "ionization potentials," more properly called "disintegration energies," representing the energies required to remove various particles completely. Finally, we can speak of quasi-stationary states. These states, however, no longer represent the motion of a single particle in a central field, but rather refer to the various possible motions of the neutrons and protons comprising the nucleus, under the influence of their mutual forces. The term "quasi-stationary" is used in all cases since even below the lowest disintegration energy, decay can usually proceed by gamma-ray emission. As far as is known, the spacing of these levels does not follow any regular pattern, except that the levels tend to become closer together at higher excitation energy and to be more closely spaced for nuclei of high atomic weight (greater number of neutrons and protons).

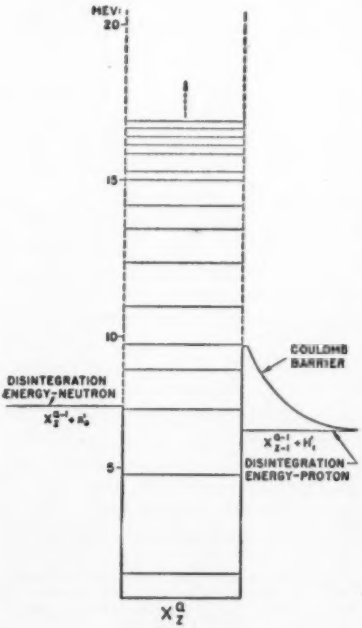

FIG. 2

The disintegration energy is the energy which must be concentrated on one particle to eject it from the nucleus and will have various values depending on which particles, or groups of particles, are to be removed. A charged particle having this amount of energy must still penetrate the electrostatic or "coulomb" barrier to escape completely, and the probability of its doing so will be very small unless considerable additional energy is available. For the neutron, no electrostatic forces are involved, so that a neutron having the disintegration energy may escape. The essential difference from the atomic case lies in the fact that the excitation energy is usually shared among all the particles and only rarely will it happen to be concentrated on any one particle. Since we are dealing with the motions of a system of several particles, the total energy can be considerably greater than the disintegration value without any single particle having sufficient energy

to escape. This fact has the consequence that quasi-stationary states of the nucleus can exist far above the disintegration energy for a sufficient length of time to permit radiation. It was the discovery of these high states through the observation of radiation accompanying the capture of protons and neutrons which first led Bohr to formulate the present picture of the nucleus.

The fact that in the nucleus we are dealing with energy differences of millions of electron volts means that the changes in the mass of the particles involved can no longer be ignored;* indeed, it is just by the measurement of the masses of the various systems that the energies involved in various reactions are predicted, and conversely, from measurements of energy released in nuclear reactions, the masses of nuclei not available in nature can be ascertained. Furthermore, the high excitation energies needed for nuclear spectroscopy are often to be attained most easily by combining particles artificially and utilizing the energy representing the change in mass. Thus one can produce a given nucleus in an excited state by adding a particle to a lighter nucleus, taking advantage of the fact that a close combination of nuclear particles represents less energy than does the sum of their individual masses. The excitation energy gained by addition of a particle to a nucleus is equal to the "binding energy" of that particle in the combination nucleus and averages, for neutrons and protons, about 7 Mev. For deuterons the binding energy may be of the order of twice this figure, while for α particles the value is considerably smaller, since the α particle is itself so tightly bound as to contribute very little excess mass for excitation. In the light elements, the binding energies vary considerably so the excitations obtained must be calculated for each individual case.

* The mass of the electron is equivalent to 0.511 Mev of energy.

Nuclear Reactions

It is convenient to describe these processes in the nomenclature of nuclear reactions in some such form as:

$$(a)\quad M_0 + M_1 + E_1/c^2 \rightarrow (M) \rightarrow M_2 + M_3 + E_2/c^2$$

$$(b)\quad M_3 \rightarrow M_4 + M_5 + E_3/c^2$$

where the events taking place can be described as follows: the nucleus M_0 collides with the particle M_1, M_0 and M_1 having a total kinetic energy E_1 (in center-of-mass coordinates); a "compound nucleus," M, is formed, having a number of neutrons and protons given by the sum of those contained in M_0 and M_1 and having an energy given by the sum of the masses plus the kinetic energy, E_1. The compound system, then, is a configuration of neutrons and protons corresponding to some nucleus, but with an amount of energy, determined by the experiment, which is in general different from that represented by any particular state. If the excitation energy coincides with that of a quasi-stationary state of the configuration, the system may have a relatively long lifetime and will behave in a manner characteristic of this state. The compound nucleus decays to the "residual" nucleus, M_3, with emission of a particle M_2, M_3 and M_2 having a total kinetic energy E_2. The nucleus, M_3 may further decay into the nucleus M_4, with emission of a particle, M_5, and energy release, E_3. As used here, M_1, M_2 and M_5 may represent any particle or radiation not prohibited by considerations of conservation laws or selection rules. In the majority of cases, M_3 is produced in the ground state or in states which decay by gamma-ray emission. In case particle emission is possible the process is often referred to as a three-body reaction. Once M_0 and M_1 are combined, M is

of course uniquely determined, but the further course of the reaction will involve competition between several possibilities, including re-emission of a particle identical to M_1 (resonance scattering). It is by the study of the probabilities of the various modes of disintegration, as influenced by the excitation energy, that information about the levels of M is obtained; observation of the radiation associated with the decay of M_2 or of apparent deficiencies in the energy release E_2 leads to information about the levels of M_2.

The existence of the compound nucleus as an intermediate stage in a nuclear reaction can be considered in a quantitative fashion by employing an argument first advanced by Bohr. It is well known that the width of the resonance levels observed in the excitation of the system represented by the number of protons and neutrons making up the compound nucleus is related to the mean lifetime of the system by the uncertainty principle. If Γ is the full width at half maximum of the resonance in energy units and τ is the mean lifetime then

$\Gamma\tau \simeq \hbar \sim 10^{-15}$ electron-volt-seconds.

Distinguishable resonances in nuclear reactions involving light nuclei have been observed having widths from about 10^6 electron volts down to values as small as 10^{-1} electron volts. These correspond to lifetimes from 10^{-20} to 10^{-14} seconds. The particles which initiate nuclear reactions and which are produced in nuclear reactions have velocities of about 10^9 cm per sec so that the times of passage through nuclear systems of the order of 10^{-13} to 10^{-12} cm in diameter should be 10^{-22} to 10^{-21} seconds in duration if only single collisions occurred and no intermediate quasi-stationary states were involved. The longer times inferred from the resonances indicate unmistakably that even in light nuclear reactions the compound nucleus has a lifetime long compared to the characteristic times for traversal of its diameter by the constituent particles. It is true that in numerous cases the excitation curves over considerable energy ranges show a slowly varying yield (on which sharp resonances are sometimes superimposed). In these cases the concept of the compound nucleus is not as useful since the rate and nature of the nuclear reaction may depend to a large extent on the method of its initiation, that is on whether protons, neutrons or other particles are employed as bombarding agents. Nevertheless, with this qualification in mind, it is useful to consider this situation as a special case in which the lifetime of the compound nucleus is relatively short. Study of these cases will certainly be important but in this discussion we are primarily concerned with those states which exist for a long enough time to be characteristic of a given number of neutrons and protons and thus to have a considerable effect on the rate and nature of a variety of nuclear reactions in which they can be produced as intermediate stages.

Bohr has emphasized that the long lifetime arises primarily from the distribution of the energy among the various constituents in the compound nucleus. A contributing factor is also the existence of certain selection rules which may prevent one or more modes of decay which are otherwise energetically possible. The first case of this type was observed in the disintegration of Li^7 by protons with gamma-ray emission. Resonance in the emission of 17.6 Mev gamma rays occurs at a bombarding energy of 440 kev with a half-width of 12 kev, corresponding to a lifetime of the intermediate nucleus Be^8 of 10^{-19} seconds. However, at this same energy, the emission of two 8.8 Mev alpha particles is observed to take place with no sign of resonance in the yield curve which varies in a smooth fashion from very low to very high

energies. This dilemma was clarified by the realization that only states of even angular momentum and with certain symmetry properties could decay with the emission of two identical particles and that states without these properties could be produced by combining Li^7 and a proton. The only modes of decay for these latter states at energies where neutron emission is energetically impossible are gamma emission or the re-emission of a proton (not necessarily the same as the incident proton). Since the protons have low energy they have difficulty repenetrating the electrostatic barrier and in the relatively long period of this re-emission (10^{-19} seconds), high-energy, gamma-ray emission (10^{-16} sec) can successfully compete in about one case in one thousand. This is enough to make the gamma radiation easily detectable and to reveal the surprising character of this particular state of Be^8. The simultaneous emission of alpha particles is of course attributable to the production with Li^7 and protons of the same energy of states which do have the required angular momentum and symmetry properties. Gamma radiation cannot compete with the alpha decay of these states and thus it occurs prolifically only when a state which cannot so decay is also produced.

The excitation of a nucleus by addition of energy through γ-radiation is represented by regarding the bombarding agent, M_1, as a γ quantum:

$$M_0 + \gamma \rightarrow (M_0) \rightarrow M_2 + M_3 + E_2$$

where the compound nucleus is identical to the target, except in energy. The decay can then proceed either by re-emission of a γ quantum, or, if sufficient energy is available, by emission of a particle. The latter process, photo-disintegration, has been observed in many elements, the most notable being the deuteron (threshold, 2.16 Mev) and Be^9 (1.63 Mev). The states involved, however, are so broad that no definite levels can be located. In the case of Be^9, disintegration by electron bombardment has also been observed; the electron in this case is not thought actually to enter the nucleus, but rather to transfer a part of its kinetic energy in the form of a "virtual" γ-quantum. No examples of the photo excitation are known in which radiation is emitted (scattering, or resonance fluorescence), although the problem of detection of such radiation in the presence of the primary beam is such as to make it unlikely that the effect would be observed unless it were very strong indeed. It might be expected that anomalous absorption of γ-radiation would be exhibited for γ-ray energies coincident with the excitation energy of stationary states in the bombarded nucleus. (Compare resonance absorption in atomic levels.) The theoretically predicted cross section is, however, very small, and the effect has not been observed.

Energy Level Diagrams

Figure 3 shows a level diagram of Li^7, including several examples illustrating the location of levels from particle reactions. Here are plotted the various combinations of target nuclei and bombarding particles which yield Li^7 as a compound nucleus, together with those combinations which leave Li^7 as a residual nucleus. Each combination contains a total of three protons and four neutrons, and the excess mass of the various configurations over the ground state mass of Li^7 is plotted vertically in millions of electron volts. Thus, the combination, $Li^6 + n$, has a total energy just 7.15 Mev greater than normal Li^7 (6.01697 + 1.00893 − 7.01822 = 7.68 millimass units = 7.15 Mev). The absorption of slow neutrons by Li^6 leads to the formation of Li^7 with 7.15 Mev of excitation. The only observed mode of disintegration, in addition to the scattering, is the decay

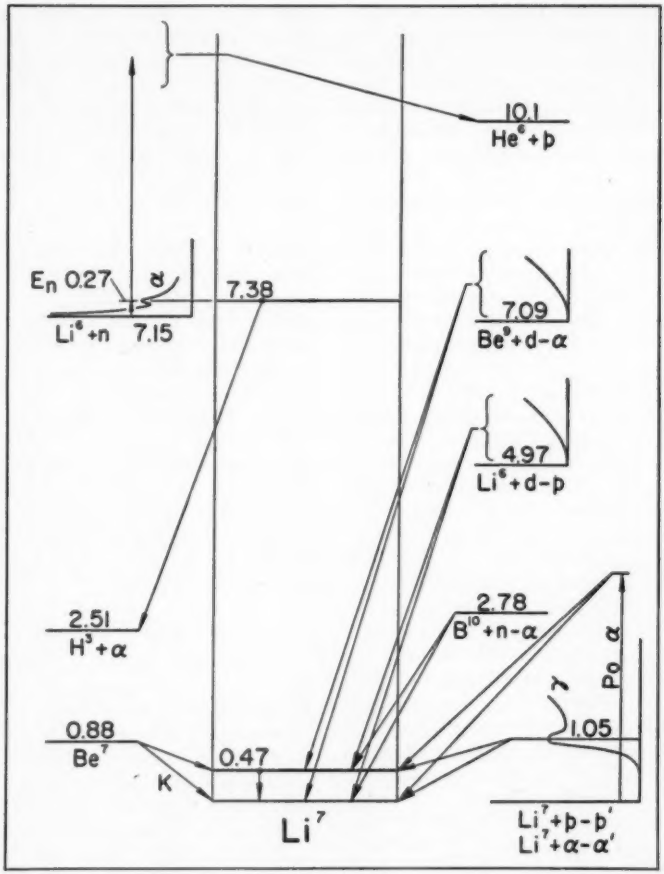

FIG. 3

into a tritium nucleus and an α particle:

$$Li^6 + n \rightarrow (Li^7) \rightarrow H^3 + He^4 + 7.15 \text{ Mev}$$

The yield of α particles as a function of neutron energy—indicated horizontally on the curve marked "α"—shows the initial decrease with increasing energy characteristic of neutron absorption, and in addition a pronounced maximum, or "resonance" for a neutron energy of 0.27 Mev. The appearance of this resonance is evidence for a quasi-stationary state of Li^7 at 7.38 Mev ($7.15 + 0.27 \times 6/7$) which disintegrates as indicated. At much higher neutron energies sufficient energy is available to permit decay into He^6 and a proton:

$$Li^6 + n \rightarrow (Li^7) \rightarrow He^6 + H^1 - 2.95 \text{ Mev}$$

Alpha and H^3 particles are presumably

also produced here, although they have not been observed. Whether well defined levels of Li^7 exist in this region is not known.

Another example of interest is the bombardment of Li^7 with protons. Here Be^8 is the compound nucleus, and one of the possible modes of disintegration, where sufficient energy is available,* leads to re-emission of a proton of lower energy, leaving Li^7, the residual nucleus, in an excited state (at ~ 0.47 Mev) which decays by γ-radiation:

$$Li^7 + H^1 \rightarrow (Be^8) \rightarrow Li^{7*} + H^1 - 0.47 \text{ Mev}$$
$$Li^{7*} \rightarrow Li^7 + \gamma \ (0.47 \text{ Mev})$$

The shape of the yield curve (marked "γ") shows initially a roughly exponential increase in the probability of this particular reaction with increasing proton energy—corresponding to the increasing ease with which the incoming and outgoing protons can penetrate the Coulomb electrostatic barrier. At a proton energy of 1.05 Mev—representing an excitation in Be^8 of 18.13 Mev—a resonance appears, indicating a relatively well-defined excited state of Be^8 leading to this mode of decay.

The same state of Li^7 can be reached by bombardment of Li^7 with polonium α-particles. Other cases of this so-called "non-capture" reaction with protons occur in B^{10} (0.420 Mev state) and Ne^{20} (1.4 Mev). There is some evidence for non-capture excitation of Be^9 by α-particles, but the states involved decay by neutron emission rather than by radiation, because the binding energy of a neutron in Be^9 is relatively small.

In the heavy elements, a great amount of information has been obtained through the study of radioactive disintegrations where the excited states of the daughter nucleus are deduced by differences in energy of the particle groups or by observation of γ-radiation accompanying the decay. This method has application to a number of the light nuclei, particularly Li^7, Be^8, C^{12}, O^{16} and Ne^{20}. In our example (Fig. 3), the nucleus Be^7 decays with a half-life of 53 days to Li^7 by capture of an extra-nuclear electron, accompanied presumably by emission of a neutrino. Two types of decay are observed; the more probable one leaves Li^7 in the excited state at ~ 0.47 Mev and is hence accompanied by γ-radiation, while the less probable mode of decay leads directly to the ground state:

$$Be^7(+e^-) \begin{cases} \rightarrow Li^7 + 0.88 \text{ Mev} \\ \rightarrow Li^{7*} + 0.41 \text{ Mev} \end{cases}$$
$$Li^{7*} \rightarrow Li^7 + \gamma \ (0.47 \text{ Mev})$$

The case of Li^8 (half-life 0.88 sec) which decays to excited states of Be^8 is considerably more complicated, but the energy distribution of the β-particles indicates that transitions to a state of Be^8 at ~ 3 Mev and possibly to higher states are involved. As far as is known, all these states decay by α-emission and are very broad, although there is some evidence for a γ-ray from a state at 7.5 Mev with subsequent α-emission from the ground state of Be^8. B^{12}, with a half-life of 0.022 sec, is another case of considerable complexity. Again, the β spectrum shows several groups, indicating formation of excited states in C^{12}, but the decay of these states has not been observed.

The production of α-particles in the bombardment of Be^9 with deuterons affords a good example of the location of energy levels in a residual nucleus by differences in the energy release, E_2. Here two groups of α particles are observed, corresponding to the two possible reactions (Fig. 3):

$$Be^9 + d \rightarrow (B^{11}) \begin{cases} \rightarrow Li^7 + \alpha + 7.09 \text{ Mev} \\ \rightarrow Li^{7*} + \alpha + 6.62 \text{ Mev} \end{cases}$$
$$Li^{7*} \rightarrow Li^7 + \gamma \ (0.47 \text{ Mev})$$

* This reaction is energetically impossible at the resonance energy (0.440 Mev) found for the production of high-energy gamma radiation which has been previously discussed.

The long-range α particle group leaves Li^7 in the ground state, while the other group, having lower energy, leaves Li^7 in the excited state at 0.47 Mev, which then decays by γ-ray emission. Two other reactions, $Li^6(d,p)Li^7$ and $B^{10}(n,\alpha)Li^7$ also exhibit particle groups and γ-radiation attributed to this level.

The Be^9 + deuteron reactions also yield information on the nucleus B^{10}, from energy differences in the neutron groups (Fig. 4):

$$Be^9 + d \rightarrow$$

$$(B^{11}) \begin{cases} \rightarrow B^{10} + n + 4.31 \text{ Mev} \\ \rightarrow B^{10*} + n \text{ (various)} \end{cases}$$

$$B^{10*} \rightarrow B^{10} + \gamma$$

The observed neutron groups correspond to levels in B^{10} at $\sim$ 0.6, (1.2?), 2.0 and 3.5 Mev. The gamma-ray spectrum contains lines at 0.42, 0.72, 1.03, 1.46 and 3.45 Mev, indicating levels at 0.42, 0.72 (probably unresolved in neutron work), 1.46 and 3.45 Mev; the 1.03-Mev line is probably the result of cascade transition from the 1.46 to the 0.42 Mev level. Obviously the agreement is not yet completely satisfactory, but at least the lower levels are fairly well established. The study of neutron groups in the light element reactions has been of particular importance because of the large number of levels which have been established. Because the neutron has no charge, transition to levels involving relatively small energy differences can be observed; whereas charged particles exhibit a pronounced preference for large jumps, since they require considerable excess energy to escape through the Coulomb barrier.

A case of some interest occurs in the production of low-energy neutrons in endothermic reactions. If Li^7 is bombarded with α particles (Fig. 4), no neutrons are energetically possible until an excitation energy of 2.78 Mev is available ($2.78 \times 11/7 = 4.4$ Mev α particles). At an energy slightly above this value, slow neutrons are produced:

$$Li^7 + \alpha \rightarrow (B^{11}) \rightarrow B^{10} + n - 2.78 \text{ Mev}$$

As the energy of the bombarding α-particles is increased, the energy of the neutrons increases, and if one uses a detector sensitive only to slow neutrons, the yield remains constant (assuming a thick target). At some energy higher than the threshold, an abrupt increase in yield will be observed, corresponding to transition into the first excited state of the residual nucleus. With this method, levels have been observed at $\sim$ 0.8, 1.3 and 2.1 Mev in B^{10}.

Numerous other examples of the detection of low-lying levels in residual nuclei by observation of energy differences in particle groups can be cited; in fact the bulk of our information in these regions is derived from experiments of this type. An equally important source of information, however, is the study of the yields of various disintegration products as a function of the excitation energy of the compound nucleus. One case of this kind was discussed in connection with the $Li^6(n,\alpha)H^3$ reaction, where a level in the Li^7 nucleus was involved. A somewhat more complex situation is encountered in the bombardment of Be^9 with protons, in which B^{10} is formed. The excitation of the compound nucleus—due to the difference in mass between ($Be^9 + p$) and B^{10} in the ground state—is 6.49 Mev. With this energy plus that contributed by the kinetic energy of the bombarding proton, several types of decay are energetically possible: disintegration by alpha particle or deuteron emission, re-emission of the proton, or radiation to lower levels of B^{10}. Of particular interest is the variation with energy of the γ-ray yield, which exhibits several striking features (Fig. 4). With a thin target, peaks occur in the yield curve for proton energies of 0.33, 0.67, 0.988, 1.077 and 2.52 Mev, corresponding to excitation of states in B^{10} at 6.78, 7.09, 7.38, 7.47 and 8.76 Mev. The levels

up to and including 7.38 Mev decay by transition to the ground state, while that at 7.47 decays first to the level at 0.72 Mev and then subsequently to ground. The difference between the two adjacent levels at 7.38 and 7.47 is also reflected in their widths, the former being 95 kev wide, and the latter only 4 kev wide. At 8.76 Mev (2.52 Mev bombarding energy) the gamma-ray energy is about 3 Mev indicating an entirely different mode of decay—perhaps by α particle emission to an excited state in Li^6 or, less probably, by multiple transitions through lower levels in B^{10}. This same level can also decay by neutron emission, as shown by the pronounced resonance in neutron yield at 2.52 Mev. The yield of α particles and deuterons has been studied up to about 1.4 Mev and the character of these curves is best explained by the assumption of very broad, poorly defined states, modified by interference with the resonance levels which appear in the yield of gamma radiation. The

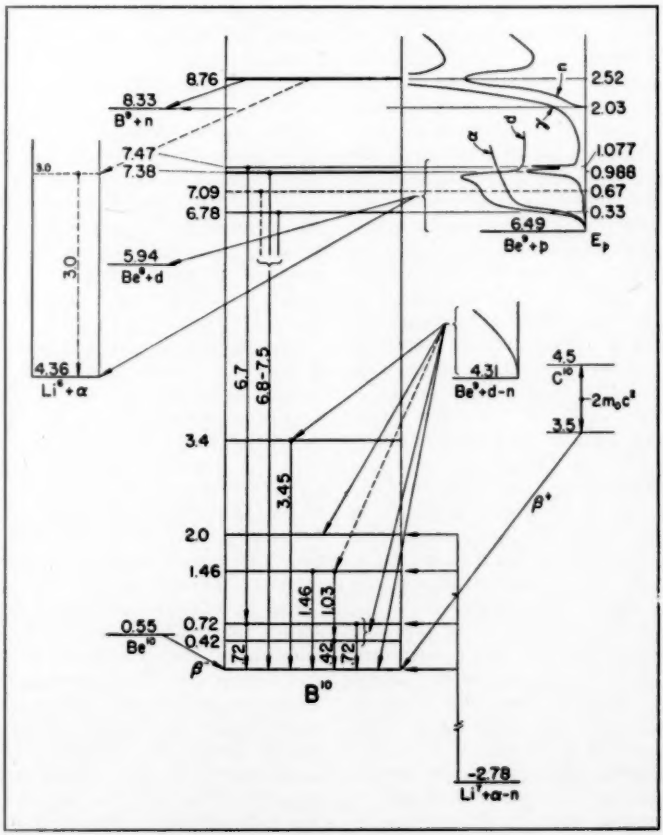

FIG. 4

same sort of behavior is exhibited by the scattered protons, which show pronounced anomalies at the gamma ray peaks at 0.988 and 1.077 Mev.

As a matter of fact, one should in principle be able to map all the excited states of a nucleus in a given region by observation of the scattering of the bombarding particle, with a few exceptions where strong selection rules prohibit interaction with particular states. In the example discussed above, one of the possible courses of the reaction is the re-emission of a proton from the compound nucleus B^{10}; this process may be regarded as the scattering of the proton by the Be^9 nucleus. The character of this scattering process will be modified to some extent at each level, whether other types of decay occur or not. When charged particles are used, the specifically nuclear scattering must be distinguished from the Rutherford scattering due to the interaction of the Coulomb forces by its dependence on energy or by the distribution as a function of angle. In the neighborhood of quasi-stationary states of the compound nucleus, the probability of scattering may exhibit a rapid variation with energy and ordinarily large angles of deflection will be relatively more probable than is the case in Rutherford scattering. Very important information concerning the character of the scattering state, for example as regards its angular momentum, can be obtained from these angular distribution studies although the interpretation of the results is not always simple.

With charged bombarding particles, the probability of forming a compound state depends markedly on the kinetic energy available, since the Coulomb barrier must be penetrated. This fact has the consequence that only strong levels will be observable for low bombarding voltages, and hence in scattering or disintegration experiments some regions will suffer from this discrimination. For kinetic energies of the order of one to three Mev for protons and twice these values for α particles, this effect becomes of minor importance in the light elements. Since sources of mono-energetic α particles up to 8 Mev are available from the naturally radioactive elements, the study of α-particle scattering was one of the earliest attacks on the problems of the light elements, and, with later work, levels have been established in Be^8, O^{16}, and Ne^{20}. Proton scattering has been extensively investigated in connection with the first few nuclei, and in the region of interest here has yielded information on Be^8, Be^9, B^{10} and Ne^{20} (scattering in Li^7, Be^9, non-capture in Ne^{20}).

The neutron offers certain advantages in the study of scattering processes, since it is not affected by the Coulomb field, and a great deal of work has been done in mapping levels by neutron scattering. Among the light elements in which levels have been located by this method might be mentioned Li^7, Be^{10}, B^{11}, C^{13} and N^{15}.

Although, as the above illustrations indicate, a considerable variety of techniques is available for the identification of nuclear levels, the regions of interest are by no means uniformly covered. The application of any particular method may be limited by the amount of excitation attainable, in the availability of a suitable target nucleus, and by the probability of occurrence of a reaction. Thus, for example, the methods of non-capture and photo-disintegration are applicable only to the stable isotopes, or to radioisotopes of relatively long life. The observation of resonances in the compound nucleus is again restricted to those nuclei which can be formed from available target nuclei and is further limited in the energy interval which can be surveyed. The bombardment of Be^9 with protons, for example, leads to excitation in

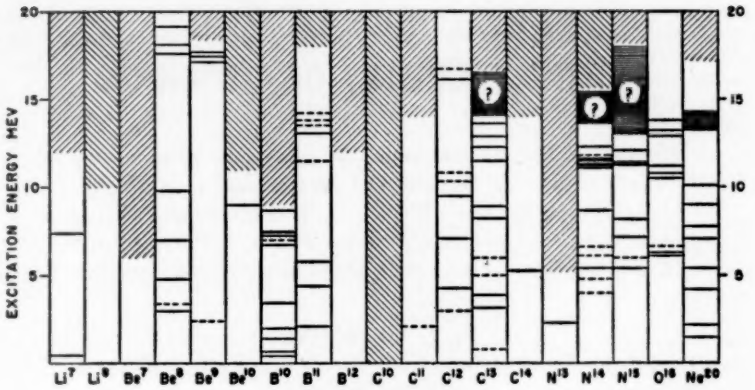

FIG. 5

B^{10} from 6.49 Mev to an upper limit depending on the proton energy available. Even the region immediately above 6.49 Mev is poorly covered because of the small probability of penetrating the Coulomb barrier with low energy protons. The detection of levels from disintegration particle groups depends on the existence of an appropriate combination of target and bombarding particle and of a sufficient probability for the particular mode of decay involved. All of those restrictions vary in a more or less random manner from one nucleus to another with the consequence that some regions have been fairly completely mapped and others not at all. The nucleus C^{10} is in a particularly ill-favored position; it cannot be formed as a compound nucleus by α-particle, proton, neutron, or deutron bombardment and is observed as a residual nucleus in only one reaction, $B^{10}(p,n)C^{10}$.

A summary of the known levels of some of the light nuclei is presented in Fig. 5. Each level is represented by a horizontal line at a height on the vertical scale indicating the excitation energy above the ground state. Levels whose existence is considered as doubtful are indicated by broken lines. The high groups in C^{13}, N^{14}, and N^{15} are reported as individual levels but may represent unresolved, broad excitation states. The incompleteness of the present information is indicated to some extent by the cross-hatching of regions which are thus far inaccessible or have not yet been investigated. It should also be pointed out that the levels plotted are largely those of small width; the entire region is presumably permeated with broad levels of undefined location which would have a bearing on any complete picture of the nucleus. Certainly it is clear that much work remains to be done before one can say whether or not the kind of regularities characterizing transitions in atomic systems will appear in the spectra of the light nuclei.

THE ACTINIDE ELEMENTS and THE CHEMISTRY OF PLUTONIUM

Review of the characterization of plutonium based on the theory of actinide elements and experimental work at Berkeley, Chicago and Chalk River. Only millionths of a gram of plutonium were available for these studies, but they formed the foundation for large-scale chemical separation operation

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AT VARIOUS TIMES it has been suggested that at least some of the elements in the last period of the Periodic System might form the beginning of a new series analogous to the rare earths. However, the physics and chemistry of the heavy elements were too obscure to enable judgment to be passed upon the suggestion.

The subject was reopened by G. T. Seaborg after a study of the properties of the four transuranic elements discovered since 1940 (*1*).

The Actinide Theory

Seaborg suggests that the elements from actinium to curium inclusive form the first part of a new rare-earth-like series, which, from the name of its first member, is known as the *Actinide Series*. The peculiar properties of the lanthanide rare earths have long been attributed to the presence of an incompleted shell of 4*f* electrons in these elements. The 4*f* shell is filled up as the series is ascended. The properties of the actinide elements are interpreted in an exactly analogous manner by assuming that these elements possess an incomplete shell of 5*f* electrons.

On the simplest assumption, the first 5*f* electron would appear in Th, and there would be a regular increase of one 5*f* electron for each unit increase in atomic number up the series. Element 96 (curium) would therefore have a $5f^7$ configuration. However, this regular addition to the *f* shell is known not to occur in the lanthanide elements (*2*), among which the configuration in which there is no 5*d* electron is commonly found, *e.g.*, Sm has $4f^6 5s^2 5p^6 6s^2$ rather than $4f^5 5s^2 5p^6 5d^1 6s^2$. It is very likely that a similar phenomenon occurs in the actinide series. The configurations in Table 1 have been suggested (*3*).

Spectroscopic. Partial analyses of the emission spectra of thorium and uranium have been published. It has been shown that no 5*f* electrons occur in

TABLE 1

Configurations of Actinide Elements

Element	*Symbol*	*At. No.*	*5f*	*6s*	*6p*	*6d*	*7s*
Actinium	Ac	89	0	2	6	1	2
Thorium	Th	90	0	2	6	2	2
Protoactinium	Pa	91	2	2	6	1	2
Uranium	U	92	3	2	6	1	2
Neptunium	Np	93	4	2	6	1	2
Plutonium	Pu	94	5	2	6	1	2
Americium	Am	95	6	2	6	1	2
Curium	Cm	96	7	2	6	1	2

the normal states of neutral or ionized Th. The following configurations have been established (*4*):

Th I $6d^2\ 7s^2$ (probable)
Th II $6d^2\ 7s^1$
Th III $6d^2$ (probable)
Th IV $6d^1$

Excited states of Th II containing *5f* electrons begin 4,490 cm^{-1} above ground level.

The following configurations have been established for uranium (*5*):

U I $5f^3\ 6d^1\ 7s^2$
U II $5f^3\ 7s^2$

What little spectroscopic evidence there is, therefore, definitely indicates that uranium is a member of a rare-earth-like series, but the possibility that the series begins with Th rather than Ac must still be entertained.

Absorption Spectra. The absorption spectra of compounds of the lanthanide rare earths are unusual for the sharpness of the absorption bands. These sharp lines are now generally attributed to electronic transitions within the *4f* shell (*2*). Since this shell is well protected by the *5s* and *5p* shells, the *4f* energy levels are not greatly perturbed by solvation or complexing of the lanthanide ions. Hence the sharpness of the absorption bands is reminiscent of the absorption spectrum of a gas rather than of a solution.

The absorption spectra of U^{4+}, the lower oxidation states of neptunium, and all the valence states of plutonium have been shown to contain sharp bands (*6,7*) which, by analogy with the lanthanide elements, are supposed to arise from transitions within the *5f* shell.

For the configurations $4f^1$, $4f^{13}$, $5f^1$ or $5f^{13}$, only one electronic transition is possible (*6*), so that the spectra of ions containing these configurations should contain only one sharp line. These configurations should be present in the ions Ce^{3+}, Np VI and Yb^{3+}. However, the spectra of these ions have been shown to contain no sharp lines at all, so that the one possible transition must either be highly forbidden, or else lie outside the spectral regions which have been investigated.

Th IV and U VI, which contain no *5f* electrons at all, give rise to no sharp absorption lines.

Paramagnetic Properties. The paramagnetic susceptibilities of the lanthanide rare earths have been calculated by Van Vleck and Frank (*8*), assuming that the tripositive ions have a $4f^n$ configuration, where *n* goes from zero to 14. Excellent agreement between the calculated and measured values of the susceptibilities was obtained, and provides one of the best pieces of evidence for the $4f^n$ configuration.

Van Vleck has also calculated the paramagnetic susceptibility of the uranous ion, but he assumed that the configuration was $6d^2$ rather than $5f^2$. The calculated values of the susceptibility are not in good agreement with experiment.

The ion Th^{4+} was found by Bose (*9*) to be diamagnetic, but this is to be expected since the ion should contain no incompleted shells, either *5f* or *6d*.

From lack of theoretical and experimental attention, therefore, the paramagnetic evidence for or against the *5f* configuration is entirely inconclusive.

Chemical Properties. The chemical properties of the actinide elements cannot provide any direct evidence for the presence of *5f* electrons in these elements. However, it has been found that, particularly in the higher members of the series, the chemical properties are consistent with a *5f* configuration.

The characteristic valency of the lanthanide rare earths is +3. The actinide elements do not exhibit such a strongly marked characteristic valency. Indeed, the valencies of Ac, Th, Pa and U appear to place them in groups 3, 4, 5 and 6 of the Periodic System rather

TABLE 2

Valencies of Lanthanide and Actinide Elements

Element	*Valencies*	*Element*	*Valencies*
La	+3	Ac	+3
Ce	+3, +4	Th	+4
Pr	+3, +4, +5	Pa	+4, +5
Nd	+3, +4 (?)	U	+3, +4, +5, +6
61	+3	Np	+3, +4, +5, +6
Sm	+2, +3	Pu	+3, +4, +5, +6
Eu	+2, +3	Am	+2, +3, +4
Gd	+3	Cm	+3

than in a rare-earth-like series. The known valence states of the actinide and lanthanide elements are shown in Table 2 (*2,10*).

Both series start with a purely trivalent element, and both have a purely trivalent element in the eighth place. Between the first and eighth members, higher valencies at first appear and then disappear again. Just before the eighth member, both series contain elements with a dispositive valency.

In order to account for the stable hexavalent states of U, Np and Pu, one must suppose that the $5f$ electrons of these elements are rather less firmly bound than the $4f$ electrons of the lanthanide rare earths. There is therefore a greater tendency for ions of higher valencies to appear.

The $4f^7$ configuration of Gd is known to be particularly stable (*2*), and it is the tendency towards the formation of this configuration which leads to the appearance of divalency in the elements just before Gd. The divalence of Am must be attributed to an analogous stability of the $5f^7$ configuration of Cm. There are other examples in the Periodic System of the stability of half-completed shells, so that the stability of the configurations of Gd and Cm is not surprising.

The chemical similarity between U, Np and Pu is particularly marked, and can only be satisfactorily accounted for by the actinide theory. This similarity, and the exclusive trivalence of curium, constitute the best chemical evidence for the theory.

The Chemistry of Plutonium

The very interesting chemical properties of plutonium have been intensively studied in the USA and Canada. The half-life of Pu^{239} is fortunately long enough to enable the chemist to handle the element without too much risk or difficulty.

Plutonium was first obtained by Seaborg, McMillan, Kennedy and Wahl (*11*) at Berkeley late in 1940. The isotope Pu^{238} was produced by the following process:

$$U^{238}(d, 2n)Np^{238} \xrightarrow[2.0\ \text{days}]{\beta} Pu^{238}(\alpha, 60\ \text{years})$$

Since 1940, large amounts of pile-produced Pu^{239}, an α-emitter with a half-life of 2.411×10^4 years (*12*), have been available for chemical studies.

Natural Occurrence. Szilard suggested that uranium minerals might contain a very small amount of Pu^{239}, which would be formed by spontaneous fission of uranium followed by capture of fission neutrons in U^{238} to give U^{239}. By two successive β-decays, U^{239} would produce Pu^{239}.

Seaborg made a very careful search for plutonium in pitchblende and was able to isolate a small amount of an α-emitter with the chemical properties of plutonium. It was decided that the concentration of plutonium in pitchblende was of the order of 1 part in 10^{14}. Assignment of observed α-radiation to Pu^{239} is perhaps not certain enough for the occurrence of plutonium in nature to be established definitely (*13*).

Chemical Properties. Valency states of +3, 4, 5 and 6 have been established. The actinide theory leads to the expectation that, in the elements beyond

uranium, the lower valency states will become progressively more stable. The most stable valency state of uranium is +6, while the lowest state, +3, is such a powerful reducing agent that it liberates hydrogen from water. The most stable valency state of plutonium is +4, but both the +6 and +3 states are readily prepared and are stable enough to be easily worked with.

The existence of the +3, +4 and +6 states was demonstrated as follows (*14*):

1. The most stable state was shown to be tetravalent by weighing a sample of its insoluble iodate and igniting it to oxide. The loss of weight agreed exactly with the reaction

$$Pu(IO_3)_4 \rightarrow PuO_2 + 2I_2 + 5O_2$$

When a sample of the oxide was fumed to dryness with sulfuric acid, the gain of weight corresponded to the reaction

$$PuO_2 + 2H_2SO_4 \rightarrow Pu(SO_4)_2 + 2H_2O$$

2. The state obtained by reduction of Pu IV was shown to be trivalent in two ways. In the first, the iodine liberated in the reaction

$$Pu\ IV + KnI' \rightarrow Pu(IV - n) + \frac{n}{2} I_2$$

was titrated with thiosulfate, and n was found to be unity.

In the second experiment, the potential of a platinum electrode was measured in solutions containing Pu IV and the lower state. The potential was found to be given by the Nernst equation

$$E = E_0 + \frac{RT}{nF} \ln \frac{[Pu\ IV]}{[Pu(IV - n)]},$$

when n, the valence difference between the upper and lower oxidation state, was set equal to unity.

3. The state obtained by the oxidation of Pu IV was found to be hexavalent by a direct permanganate titration of $Pu(SO_4)_2$ in warm acid solution.

The pentavalent state of plutonium in solution has been studied by Kasha and Sheline (*15*). They found that Pu III, Pu IV, Pu V and Pu VI could exist together in equilibrium in hydrochloric acid solutions. Normally the concentration of Pu V at equilibrium was very small, but in very dilute hydrochloric acid, up to 35 % of the plutonium in the solution was present in the pentavalent form, and the concentration of Pu IV was very small. The concentrations of all the ionic species present could be estimated by means of absorption spectrum measurements, and the kinetics and equilibria in this complicated system were studied in some detail.

The equilibrium

$$3Pu\ IV \rightleftharpoons 2Pu\ III + Pu\ VI$$

was studied. The disproportionation constant

$$K = \frac{[Pu\ III]^2[Pu\ VI]}{[Pu\ IV]^3}$$

was found to have the values 1×10^{-4} in 1.545 M hydrochloric acid and 6.9 in 0.183 M hydrochloric acid, both at 25° C. K was inversely proportional to the 5th power of the acid concentration. At 70° C, K was 300 — 4000 times greater than at 25° C, depending on the acid concentration.

At lower concentrations of hydrochloric acid, it was found that considerable quantities of Pu V appeared. In order to avoid the formation of colloidal Pu IV at these low acidities, mixtures of Pu III and Pu VI were made in concentrations of hydrochloric acid down to 0.06 M. For the reaction

$$Pu\ III + 2\ Pu\ VI \rightleftharpoons 3\ Pu\ V,$$

the equilibrium constant

$$K = \frac{[Pu\ III][Pu\ VI]^3}{[Pu\ V]^3}$$

was found to have the values 8.1 and 0.59 in 0.1081 M and 0.0615 M hydrochloric acid, respectively. The concentration of Pu IV under these conditions was quite small.

Pentavalent plutonium is supposed to exist in solution as the oxy-ion PuO_2^+,

which differs from the hexavalent oxy-ion PuO_2^{++} only in charge. The pentavalent ion is therefore analogous to that of uranium, which has been shown (*16*) to differ from the hexavalent oxy-ion of uranium only in charge. However, the ion UO_2^+ appears to be much less stable than PuO_2^+, and only exceedingly low concentrations of it normally exist in solution in equilibrium with UO_2^{++}.

Trivalent Plutonium. Reduction of tetravalent plutonium with mild reducing agents gives beautiful blue solutions containing the ion Pu^{+3}. The reduction may be carried out with SO_2, hydroxylamine, the uranous ion, mercury in chloride solutions, electrolytically, or with gaseous hydrogen at a platinum catalyst.

Trivalent plutonium does not reduce water to hydrogen, and although it is slowly oxidized by air, the reaction is much slower than the corresponding aerial oxidation of trivalent uranium. The insoluble hydroxide, $Pu(OH)_3$, however, absorbs oxygen rapidly. The stability of Pu III to oxidation therefore resembles that of the ferrous ion.

Pu III may be coprecipitated from acid aqueous solutions on a carrier of lanthanum fluoride or lanthanum oxalate. The fluoride gives much more complete precipitation than the oxalate. Addition of oxalic acid to solutions containing Pu III precipitates dark, blue-green anisotropic crystals of an insoluble oxalate (*17*).

Pu III is not precipitated by such characteristic reagents for tetravalent ions as phenylarsonic acid or picrolonic acid (*14*). It is carried down by precipitates of BiOCl (*14*) or $Cb_2O_5 \cdot xH_2O$ (*17*) much less completely than is the Pu IV ion. Presumably, therefore, the basic strength of Pu III is greater than that of Pu IV.

The bright blue sulfate of Pu III crystallizes well from sulfuric acid solutions. Solutions of Pu III sulfate containing alkali sulfates give precipitates of crystalline double salts on the addition of ethanol. These compounds have the general formula $RPu(SO_4)_2 \cdot 4H_2O$, where R may be Cs, Rb or NH_4 (*17,18*). The Cs and Rb salts are lavender blue in color, but the ammonium salt is a somewhat more intense pale blue. In the absence of hydrogen, the double salts oxidize from Pu III to Pu IV at the rate of approximately 3% per hour. Analogous compounds such as $NH_4La(SO_4)_2 \cdot 4H_2O$ exist in the lanthanide rare-earth series.

More complicated double salts of the type $R_4Pu(SO_4)_4$, where R is a potassium or thallous ion, have been prepared. Their properties have not yet been described. No compounds of the type $R_3Pu(SO_4)_3$, which would be analogous to such compounds as $R_3Ce(SO_4)_3$, have yet been made (*18*).

Pu III may be oxidized to Pu IV by mild oxidizing agents such as oxygen, or slowly by chlorine in acid solution (*19*). It is oxidized by nitrate ions on heating, or by concentrated nitric acid at room temperature. The oxidation by nitrate ions appears to be autocatalytic. It probably takes place by the following mechanism (*20*):

1. Initially—$2Pu^{+3} + NO'_3 + 2H^+ \rightarrow 2Pu^{+4} + NO'_2 + H_2O$. NO'_2 then reacts with more Pu^{+3}—

2. $Pu^{+3} + NO'_2 + 2H^+ \rightarrow Pu^{+4} + NO + H_2O$. More NO'_2 is then formed by the reaction—

3. $2NO + NO'_3 + H^+ + H_2O \rightarrow 3HNO_2$. It is uncertain whether reaction 2 or 3 is the rate-controlling step.

Tetravalent Plutonium. The only oxide of plutonium which has been described is PuO_2 (*14*). It may be prepared by the ignition of the hydroxide, nitrate or iodate of tetravalent plutonium at a dull red heat. It is a dark-colored substance which may be re-dissolved slowly in acids to give salts of Pu IV. If it has been ignited to a high temperature, it is difficult to dis-

solve, and fuming with sulfuric acid or even fusion with bisulfate may be necessary. When dissolved in nitric acid, $Pu(NO_3)_4$ is obtained, whereas when UO_2 is dissolved in nitric acid, the uranium is oxidized to the hexavalent state.

The soluble salts of Pu IV give red-brown solutions in water, except the nitrate, whose solutions are yellowish-green in dilute nitric acid and bright green in the concentrated acid.

Pu IV gives an insoluble hydroxide, fluoride, oxalate, peroxide and iodate, and is precipitated by such characteristic reagents for tetravalent ions as phenylarsonic acid and picrolonic acid. The soluble salts of Pu IV are very readily hydrolyzed in aqueous solution with the formation of colloidal Pu IV, which then adsorbs readily on to glass surfaces. Appreciable adsorption onto glassware occurs in tracer solutions, even in 0.02 M HNO_3. The Pu exhibits radio-colloidal behavior, and may be partly centrifuged from solution (*21*).

In the tetravalent state, plutonium complexes very readily with almost all the common anions of the strong acids. This behavior might be expected from an ion bearing four positive charges. Oxalate and carbonate complexes are inferred from the solubility of Pu IV oxalate in excess ammonium oxalate solution, and from the solubility of Pu IV phenylarsonate in ammonium carbonate solution (*14*). In the presence of ammonium thiocyanate, Pu IV is not coprecipitated on a carrier of zirconium phenylarsonate, so that it seems likely that a soluble complex thiocyanate is formed (*14*).

The reaction of Pu IV with hydrogen peroxide is particularly interesting. In not-too-acid solutions, addition of hydrogen peroxide to Pu IV produces first a red-brown color, and then a green flocculent precipitate of a peroxide. The compound, when precipitated from sulfate solutions, probably has the composition $Pu_2O_7 \cdot xSO_4$ (x = about $\frac{2}{3}$). On standing at room temperature, the peroxide decomposes and changes in appearance. The compound $PuO_3 \cdot \frac{1}{3} SO_4$ is produced. Pu therefore seems to resemble Th, which forms the peroxides $Th_2O_7 \cdot SO_4$ and ThO_3 (*22*), the latter being obtained by decomposition of the former at room temperature.

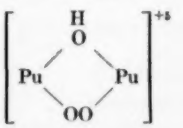
Hexavalent plutonium is reduced by hydrogen peroxide to Pu V in 0.5 M hydrochloric acid. The Pu V then disproportionates to Pu IV and Pu VI. Pu III is also formed, both by the reduction of the Pu IV by hydrogen peroxide and by the disproportionation of Pu V. In 0.5 M HCl, Pu III is the main product of the reaction. However, hydrogen peroxide also oxidizes Pu III to Pu IV, so that at equilibrium, the net result of the reaction is the catalytic reduction of hydrogen peroxide.

If only very small amounts of H_2O_2 are added to a solution containing Pu IV, no peroxide is precipitated, but first a brown complex is formed, and then, on the addition of more H_2O_2, a red complex. By the measurement of absorption spectra, the equilibria involved in this complicated system have been investigated. The following alternative structures for the brown and red complexes have been proposed (*23*):

Brown Complex:

$$Pu—OO—Pu—OH^{+5}$$

or



Red Complex:

$$HO—Pu—OO—Pu—OOH^{+5}$$

or

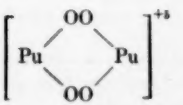


TABLE 3

Reactions of Plutonium with Some Organic Reagents

Ion	*Reagent*	*pH Range*	*Reaction*
Pu IV	Acetylacetone	2–10	Complex extractable in benzene
Pu IV	Benzoylacetone	3	Complex extractable in benzene
Pu IV	Monofluoracetylacetone	3	Complex extractable in benzene
Pu IV	Trifluoracetylacetone	2–4	Complex extractable in benzene
Pu IV	*p*-Dimethylaminobenzene-azophenylarsonic acid	4	Orange precipitate
Pu III	*p*-Dimethylaminobenzene-azophenylarsonic acid	4	No precipitate
Pu IV	*p*-Dimethylaminobenzene-azophenylarsinic acid	4	Orange-red precipitate
Pu III	*p*-Dimethylaminobenzene-azophenylarsinic acid	4	No precipitate
Pu IV	3-Nitro-4-hydroxyphenyl-arsonic acid	2.5	Pale greenish-buff precipitate. Pu carried down on Zr salt
Pu III	3-Nitro-4-hydroxyphenyl-arsonic acid	2.5	No precipitate. Pu not carried down on Zr salt
Pu IV	*m*-Nitrophenylarsonic acid	2.5	Pale greenish-buff ppt. Pu carried down on Zr salt
Pu III	*m*-Nitrophenylarsonic acid	2.5	No ppt. Pu not carried down on Zr salt
Pu IV	Phenylarsonic acid	2	Pale greenish-buff ppt. Pu carried down on Zr salt
Pu III	Phenylarsonic acid	2	No ppt. Pu not carried down on Zr salt
Pu IV	*n*-Propylarsonic acid	2	Pale greenish-buff ppt. Pu carried down on Zr salt
Pu III	*n*-Propylarsonic acid	2	No ppt. Pu not carried down on Zr salt
Pu IV	*m*-Nitrobenzoic acid	1.5	Pale greenish-buff ppt. Pu carried down on Zr salt
Pu III	*m*-Nitrobenzoic acid	1.5	No ppt. Pu not carried down on Zr salt
Pu IV	Sebacic acid	1–2	Pu carried down on Zr salt
Pu III	Sebacic acid	1–2	Pu poorly carried down on Zr salt
Pu IV	5-*p*-Acetamidophenylazo-8-hydroxyquinoline	4	Complex extractable in amyl acetate. Insol. in water
Pu IV	8-Hydroxyquinoline	4–8	Purple-brown ppt. extractable in amyl acetate

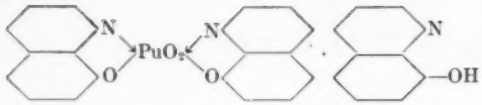
No evidence was obtained for the existence of any species containing only one plutonium atom.

The reactions of Pu with some of the more common organic reagents have been studied (*14*). See Table 3.

TABLE 3 (Continued)

Ion	*Reagent*	*pH Range*	*Reaction*
Pu VI	8-Hydroxyquinoline	4–8	Orange-brown ppt. extractable in amyl acetate
Pu IV	1′-Sulphonaphthalene-4′-azo-5:8-dihydroxyquinoline	3	Purple precipitate
Pu IV	Anthranilic acid	3	Complex extractable in amyl acetate
Pu IV	Cinnamic acid	2.5–4.5	Complex extractable in amyl alcohol
Pu IV	Cresotinic acid	2.5–4.5	Complex extractable in amyl acetate
Pu IV	2:4-Dihydroxybenzoic acid	3	Complex extractable in methyl isobutyl ketone
Pu IV	2:4-Dinitrosalicylic acid	3	Complex extractable in amyl acetate
Pu IV	Salicylic acid	2–3	Complex extractable in amyl acetate. Pu carried down on Th, La or Zr salts
Pu IV	Benzoic acid	2	Green-yellow ppt.
Pu IV	Citric acid	1–11	Soluble complex
Pu IV	Dihydroxytartaric acid	1	Pu carried down on Th salt
Pu III	Dihydroxytartaric acid	1	Pu poorly carried down on Th salt
Pu IV	3:5-Dinitrobenzoic acid	3	Complex extractable in amyl acetate
Pu IV	Phthalic acid	2	Pu carried down on Zr salt
Pu III	Phthalic acid	2	Pu not carried down on Zr salt
Pu IV	Tartaric acid	1–11	Soluble complex
Pu VI	Tartaric acid	1–11	Soluble complex
Pu IV	Thiobarbituric acid	2	Pu carried down on Th, Zr and La salts
Pu III	Thiobarbituric acid	3	Pu poorly carried down on Th, Zr, and La salts
Pu IV	Cupferron	0.5–2	Complex extractable in chloroform or carbon tetrachloride
Pu IV	Ethylenediamine	—	Brown ppt. insol. in water and organic liquids
Pu IV	α-Nitroso-β-naphthol	2	Complex extractable in methyl isobutyl ketone
Pu IV	Sodium benzenesulfinate	2	Buff ppt. not extractable in amylr acetate
Pu VI	Sodium benzenesulfinate	2	Complex extractable in amyl acetate
Pu VI	Sodium diethyldithiocarbamate	3	Purple-brown complex extractable in amyl acetate and amyl alcohol

Hexavalent Plutonium. The chemical similarity between hexavalent uranium and hexavalent plutonium suggests strongly that the ion PuO_2^{++}, analogous to the uranyl ion, exists in aqueous solutions containing Pu VI.



Requires 33.9 % Pu
Found 33.4 % Pu

Pu VI may be precipitated as sodium plutonyl acetate, either alone or on a carrier of sodium uranyl acetate ($NaUO_2Ac_3$). The crystals of sodium plutonyl acetate are tetrahedral, similar to those of sodium uranyl acetate, and the two compounds form mixed crystals (*24*).

An analysis of plutonyl oxinate showed that it has the structure shown above (*14*). This structure is exactly analogous to that of the corresponding uranyl oxinate.

The absorption spectrum of the uranyl ion contains a well-defined band system which is attributed to vibration of the U—O bond of the uranyl oxy-ion. A similar, regularly spaced system of bands has been found in the absorption spectrum of perchloric and nitric acid solutions containing the plutonyl ion (*6,7*). It seems very probable that the bands should be attributed to a Pu—O bond vibration.

Solutions containing hexavalent plutonium are slowly reduced by the products of their own α-radiation. It has been found that Pu IV appears spontaneously in a solution of pure Pu VI in 0.5 M hydrochloric acid (*15*). The reaction rate corresponds to a reduction in oxidation number of 0.0040 per day at room temperature. The rate of reduction is somewhat higher in lower concentrations of hydrochloric acid. In perchloric acid solutions, the reduction in average oxidation number is greater still—0.012 per day—and independent of the acid concentration in the range 0.1–2 M. Reduction rate is of zero order with respect to Pu VI concentration, but first order with respect to total plutonium concentration.

The addition of alkalis to solutions containing Pu VI produces precipitates of the sparingly soluble plutonates, which are analogous to the uranates. Since sodium and ammonium plutonates are slightly soluble, the precipitation of Pu VI as barium plutonate has been recommended (*25*). The solubility of this compound 15 minutes after precipitation was found to be 35 mg Pu per liter in a solution 0.25 M in Ba^{++} and 0.1 M in OH^-. After aging the precipitate for 14 hours, the solubility had fallen to 12 mg Pu per liter.

Hexavalent plutonium may be prepared by the oxidation of Pu IV with $Cr_2O_7^{--}$, $S_2O_8^{--}$, BrO_3^- in nitric acid, MnO_4^-, Ce^{4+}, or AgO. Perchloric acid at temperatures above about 170° C oxidizes Pu IV completely and rapidly to Pu VI (*26*).

Electrochemistry of Plutonium

The existence of trivalent plutonium was first recognized by the appearance of a polarographic wave in a solution of Pu IV in sulfuric acid. The half-wave potential was found to be −0.7 volts on the hydrogen scale (*14*). The molal oxidation-reduction potentials of the Pu^{3+}-Pu^{4+} couple in solutions in different acids have since been extensively studied. The results shown in Table 4, all on the hydrogen scale, have been reported.

It does not appear to be possible to oxidize Pu IV to Pu VI reversibly at an electrode. However, in perchloric acid solution, the reduction of PuO_2^{++} was found to occur at a potential very close to that of the Pu^{3+}-Pu^{4+} couple in the same acid. In 0.5 M hydrochloric acid, the potential of the Pu IV-Pu VI

TABLE 4

Molal Oxidation-Reduction Potentials of Pu^{3+}-Pu^{4+} Couple

Acid	*Potential, Volts*	*Temp. °C*	*Experimental Method*	*Reference*
1 N H_2SO_4	−0.72	23	Direct potential measurement, Pt electrode	14
1 N HCl	−0.99	30	Polarograph	14
0.5 N HCl	−0.967	?	?	20
1 N $HClO_4$	−0.86	23	Polarograph	14

couple is −1.006 volts (*20*). This potential is very near to that of the Pu III-Pu IV couple in 0.5 M hydrochloric acid. It is because the potentials of the two couples are so nearly equal that all the valency states of plutonium can exist together in appreciable concentrations at equilibrium in solutions.

The different oxidation-reduction potentials obtained for the Pu III-Pu IV couple in different acids suggest that in some of the acids, at least, some complexing with the anion must occur. *A priori*, complexing in perchloric acid would seem to be the least probable. The formation of a complex between Pu III and the chloride ion, and between Pu IV and the sulfate ion would then account for the observed potentials in hydrochloric acid and sulfuric acid. However, evidence from other sources is conflicting. The absorption spectrum of Pu IV in sulfuric acid and nitric acid suggests that the Pu IV ion is complexed in both cases (*7*). Pu III and Pu VI in 0.1 M $HClO_4$, and Pu IV and Pu V in 0.5 M HCl are said to be essentially uncomplexed (*6*), and so is Pu VI in 0.5 M HNO_3. However, it has been reported that at least two chloride complexes of Pu IV occur in hydrochloric acid solutions at room temperature (*15*).

Analytical Determination of Plutonium

The half-life of plutonium-239 is 2.411×10^4 years (*12*). One mg of Pu^{239} therefore produces 138×10^6 α-particles per minute. Absolute determination of Pu can be made in a calibrated α-counter or electroscope.

Pu may be weighed as PuO_2, $Pu(SO_4)_2$, or $Pu(IO_3)_4$ (*14*). PuO_2 may be obtained by ignition of $Pu(OH)_4$, $Pu(NO_3)_4$, or $Pu(IO_3)_4$ at red heat. The anhydrous sulfate is obtained by fuming compounds such as $Pu(OH)_4$ to dryness with sulfuric acid. The iodate may be precipitated by adding potassium iodate to solutions containing Pu IV.

The oxidation of Pu IV to Pu VI with permanganate in warm acid solution may be used for the volumetric determination of Pu. Alternatively, plutonium may be reduced with zinc amalgam in an atmosphere of carbon dioxide to the trivalent state in sulfuric acid solution and then titrated with ceric sulfate, using a potentiometric endpoint (*27*).

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ELECTRONICS FOR THE NUCLEAR PHYSICIST—III

Counting-rate meters, circuits for observing random pulses, and delay-line pulse shaping are discussed in this third of a series of four articles on fast electronic circuits, based on instrumentation techniques developed in recent years

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IX. A Counting-rate Meter

COUNTING-RATE METERS are often used in survey instruments, and probably should find more use in the laboratory to replace or to supplement the customary scaling circuit. To reassure the user of such a device, it is possible to install an auxiliary circuit controlled by a push button to supply pulses at a known rate derived from the 60-cycle supply. The calibration of the meter can thus be checked when important data are being taken. The theory of the counting-rate meter will be explained with reference to Fig. 33.

Each pulse from a conventional discriminator is supposed to trigger a circuit producing the standard rectangular pulse E shown at the left in Fig. 33. Although the height E of the pulses must be constant, the duration T is not as critical so long as the capacitor C_1 becomes fully charged before the pulse ends. If n is the counting rate, it is assumed that

$$\left.\begin{aligned} 1/n \gg T > 5R_1C_1 \\ E \gg E_0 > E_B \\ C_2 \gg C_1 \end{aligned}\right\} \qquad (67)$$

In such a case each pulse deposits a charge $q = C_1E$ on C_2, so the average potential E_0 developed with a counting rate n is

$$E_0 = nqR_2 = nC_1ER_2 \qquad (68)$$

The scale of the meter can therefore be changed by switching in accurately measured resistances R_2. The small negative bias E_B is required to keep the scale linear over the entire range of output voltage (a few volts at most). Without this bias, trouble may be encountered with the contact difference-in-potential of the diodes.

Each pulse counted excites the transient

$$e(t) = (C_1/C_2)E \exp(-t/R_2C_2) \qquad (69)$$

across C_2. The mean square fluctuation voltage for random counts can be computed using Campbell's theorem,

$$(\Delta E_0)^2 = \overline{[e(t) - E_0]^2} = n\int_{-\infty}^{\infty} e^2(t)dt \qquad (70)$$

giving

$$(\Delta E_0)^2 = \frac{nC_1^2E^2R_2}{2C_2} \qquad (71)$$

and leading to the fractional *probable*

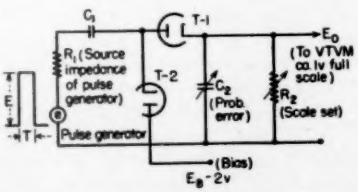

FIG. 33. Basic circuit of a counting-rate meter

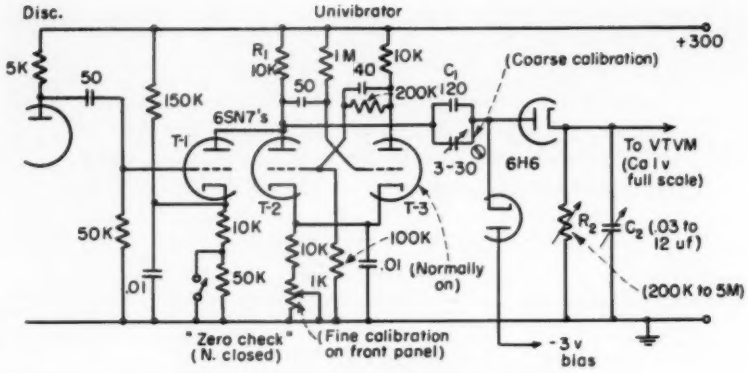

FIG. 34. Portion of a counting-rate meter

error

$$\epsilon = 0.67\frac{\Delta E_0}{E_0} = \frac{0.67}{\sqrt{2nR_2C_2}} \quad (72)$$

For a given reading of the meter, Eq. (68) shows that nR_2 is constant, so a certain value of C_2 corresponds to a definite fractional probable error of the full-scale reading.

The pulse-generating portion of a typical counting-rate meter is shown in Fig. 34 (*10*). Each positive trigger pulse from the discriminator causes a *negative* rectangular pulse to be generated at the plate of T-2 (lasting about 15 μsec). Since the time constant R_1C_1 is about 1.5 μsec, and the amplitude of the swing of the plate of T-2 is over 100 volts, the conditions (67) are satisfied. The resistors R_2 can be chosen to cover the range, for example, from 2000 to 50,000 counts per min. and the capacitors C_2 to provide probable errors of 1, 2, 5, 10, 20 percent of full scale.

Other, less complicated, but somewhat less stable counting-rate meters can be based on the use of a biased-off pentode to supply charge to the capacitor C_2 in Fig. 33. Each input pulse causes a standard pulse to be applied to the grid of the pentode, thus delivering a definite quantity of charge to the averaging circuit.

X. Circuits for Observing Random Pulses (*11*)

A cathode-ray oscillograph, together with a slave sweep circuit and an amplifier incorporating a high-fidelity delay line, enables individual pulses from a pulse amplifier to be examined in their entirety, regardless of whether the pulses occur regularly, or at random. A block diagram of a typical set-up is shown in Fig. 35. The individual portions in the set-up play the following roles.

The discriminator, which can be a simpler version of the circuit of Fig. 26,* serves to select for observation only those pulses whose amplitude is sufficiently great. In particular, it prevents the slave sweep circuit from being triggered by noise pulses.

The sweep circuit, when triggered by a pulse from the discriminator, generates a single linear sweep, of known sweep speed, and simultaneously gen-

* NUCLEONICS, Vol. 2, No. 3, 29 (1948)

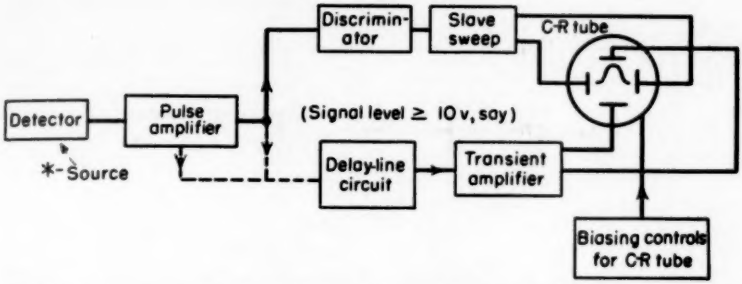

FIG. 35. Arrangement for observing the shape of random pulses with a C-R tube

erates a positive intensifying gate for the duration of the sweep. Since, for good focus, it is usually necessary to couple the sweep circuit capacitively to the deflection plates of the C-R tube, the sweep circuit should contain diode restorers to insure that each sweep shall start from the same origin. A convenient range of sweep speeds is 0.3, 1, 3, 10, etc., μsec per in. It would appear that a continuously variable sweep speed adjustment is distinctly disadvantageous, unless a very convenient means is at all times available for calibrating the sweep. Such a provision adds unnecessary complications to the circuit and its use cannot be ordinarily justified.

The delay-line circuit is based on a high-fidelity delay line. (Delay lines will be discussed in a later section.) Since most delay lines which have suitable properties possess a low characteristic impedance—1000 ohms or less—it is practicable to drive the delay line only at a few volts level. The delay-line circuit must therefore contain a means for attenuating pulses from the pulse amplifier, without introducing distortion due to stray capacitance or inductance in the attenuator. An alternative scheme involves connecting the delay-line circuit to a point in the pulse amplifier where the signal level is at most a few volts. These two methods are indicated by dashed lines in Fig. 35.

The deflection factor of a 5CP1 (or the newer 5CP1A) cathode-ray tube in an ordinary portable oscillograph is about 45 v per in. If the overall accelerating voltage is increased to 6 kv, to enable fast single pulses to be observed with ease, the deflection factor becomes about 110 v per in. In either case a transient amplifier is required between the delay line and the vertical deflecting plates. For good focus, and for convenience in obtaining large deflections, the amplifier should have a push-pull output. For ease in obtaining good response to fast transients, the amplifier should be built adjacent to the socket of the C-R tube.

The type 5CP1 C-R tube is best for visual observation of pulses whose rise time is, say, 0.1 μsec or longer. For photography of individual pulses, the 5CP11 should be substituted. With this tube operated at 6 kv, a writing speed of about 15 in. per μsec can be photographed using a fast lens and fast film. Another tube, useful where it is important to keep capacitances at a minimum (to achieve a shorter amplifier rise time), is the 5JP11. (The connections to the deflecting plates of the 5JP11 are brought out along the neck of the tube.) The focussing of this tube, however, is inferior

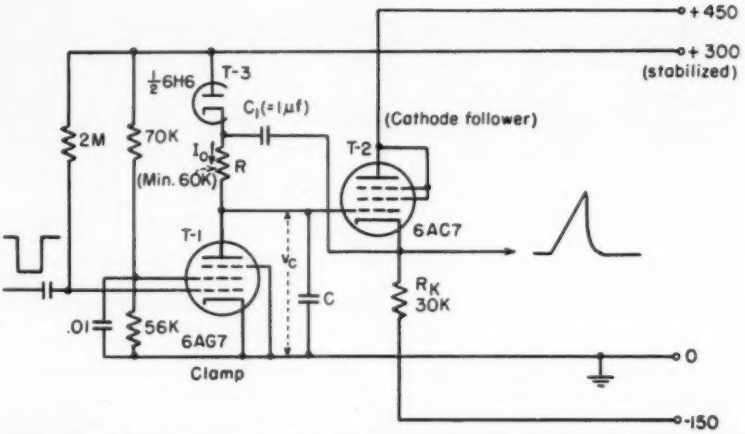

FIG. 36. Sawtooth generator of the feedback type. The sweep speed is controlled by varying C and R in steps

to that of the 5CP11. Where the utmost in performance is required, the 5RP11 can be used. This tube, operated at the maximum potential of 35 kv, enables a trace having a writing speed of about 200 in. per μsec to be photographed. The problem of designing an amplifier and a sweep circuit fast enough to justify such a writing speed is, to say the least, a formidable one.

1. Linear Sweep Generators. The obvious method of producing an approximately linearly increasing voltage for an oscillograph sweep consists in charging a capacitor with a constant current. The rate of voltage rise is then given by $dV/dt = i/C$. For example, $i = 5$ ma and $C = 100$ μμf, then $dV/dt = 50$ volts per μsec, corresponding to about one inch per μsec for the usual 5-in. portable oscillograph. To achieve a high degree of linearity, it is necessary to maintain the charging current almost constant in the face of an increasing voltage across the capacitor. In practice the sweep voltage is usually the initial part of a transient which approaches exponentially an actual, or a virtual maximum. If this maximum is the positive supply voltage —say +300 v—then it is necessary to amplify the signal to obtain a sufficiently linear voltage swing. This procedure is the one adopted in thyratron sweep generators, such as used in modern commercial oscillographs.

Another common expedient for achieving a linear sweep is to use a pentode as a constant current source for charging the capacitor. Although good linearity can be obtained with this method, the most successful sweep generators are now based on feedback arrangements, and it is a circuit of this type which will be described here. The basic circuit is one which was much used at the MIT Radiation Laboratory.

The circuit in Fig. 36 shows the essential features of a sawtooth generator of the feedback type. In the absence of a negative gating signal applied to the grid of *T*-1, current flows through the diode *T*-3, the resistance R and the pentode clamp, *T*-1. The plate of *T*-1 then rests at perhaps +10 or 15 volts. When the clamp *T*-1 is cut off by a negative gating signal, the current

through R transfers to the capacitor C, causing the grid potential of the cathode follower *T*-2 to increase. The cathode "follows" the grid and supplies, at low impedance, the output signal. In addition the output signal is fed back to the top of the resistor R. Since the top end is thus made to rise in potential almost as much as the grid end (the diode serving to disconnect the top end from the supply bus), the current through R is maintained almost constant, resulting in an almost linear sweep. The necessary charge, of course, comes from the capacitor C_1, whose magnitude is chosen to suit the greatest sweep duration. A simple analysis shows that charging current is

$$I = I_0 - v_c/FR \qquad (73)$$

where v_c is the voltage developed across C and

$$F \equiv \frac{1}{\left(1 + \frac{C}{C_1} - A\right)}, \qquad (74)$$

where A is the gain of the cathode follower. In practice F can have a value in the range 10 to 25, depending on the circuit parameters chosen. The voltage v_c is found to be

$$v_c = \left(\frac{I_0}{C}\right) t \left[1 - \frac{1}{2F}\left(\frac{t}{RC}\right) + \cdots\right] \qquad (75)$$

from which it is possible to estimate the departure from linearity. The output signal is v_c times the gain of the cathode follower (which, of course, is somewhat less than unity).

It will be noted that the plate of the cathode follower is connected to +450 v, which can be obtained from the input side of the voltage stabilizing circuit in the power supply. This device increases the permissible voltage swing of the sweep, without hurting the stability of the circuit very much. Sweep speeds are changed by switching in selected values of C and R with a two-section rotary switch. The capacitor C_1 should be at least 10 times the largest value of C used (to keep C/C_1 in Eq. (74) sufficiently small).

The recovery time of the circuit is set primarily by the time required for C_1 to lose the charge it acquires during the sweep. The order of magnitude of this time can be estimated to be $(I_0T)/(0.006)$ where I_0 is the current through R, T the duration of the sweep, and 0.006 amp the approximate current through R_K = 30 K during the recovery of the circuit. If I_0 = 2 ma, T = 100 μsec, then the recovery time is about 30 μsec. The duty ratio (fraction of time spent by the circuit producing sweeps) evidently should not be much more than 0.5, *i.e.*, the circuit is most useful in applications where it can spend a considerable fraction of its time waiting for the next trigger signal. This is no disadvantage in the majority of applications of interest to nuclear physicists.

The inverting stage shown in Fig. 37, employs voltage feedback to give good stability and linearity, and is suitable for inverting the positive sawtooth for application to the second horizontal deflecting plate of the C-R tube. The amplifier has a direct-coupled voltage feedback network from plate to grid. By properly adjusting the small trimmer capacitor C the circuit will invert fast sweeps, as well as very slow ones. On account of the considerable degeneration existing in the circuit (a gain of about 200 is fed back to a gain of unity), it is satisfactory to use the unstabilized side of the power supply

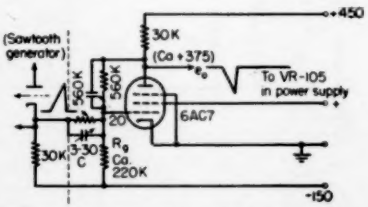

FIG. 37. Inverting stage for sweep circuit

for plate voltage. If then the plate is biased at about +375 v by choosing R_g correctly, a negative output signal of over 200 v is available without grid current being drawn, and with excellent linearity of operation.

The most useful type of gating circuit is one which automatically resets when the sweep has reached a prescribed length. This mode of operation can be realized by using a portion of the positive sawtooth, brought from a point on the resistor R_K in Fig. 36, to make a flip-flop return to the state it possessed prior to being triggered at the onset of the sweep. A satisfactory circuit of this sort is shown in Fig. 38.

Tubes *T*-2 and the cathode-grid-screen portion of *T*-3 constitute a flip-flop having two stable states with *T*-3 normally conducting with some grid current being drawn. A fast positive trigger supplied the grid of *T*-1 (normally biased off) caused the flip-flop to pass to its other stable state. Evidently the grid of *T*-3 is driven negative very rapidly (0.1 μsec) if the trigger is fast, so a fast positive intensifier gate is developed at the plate of *T*-3 and coupled out with the cathode follower *T*-5. The grid of *T*-3 is directly coupled to the grid of *T*-1 in Fig. 36 (with the capacitor and resistor shown there omitted), so the sweep starts almost simultaneously with the intensifier gate. The plate of the diode *T*-4 is direct-coupled to a point on R_K where the d-c level is considerably negative. As the sweep progresses, this point rises in potential until the diode conducts. The grid of *T*-3 is then carried into the region where *T*-3 conducts, thereby causing the flip-flop to return to its initial state, terminating both the sweep and the intensifier gate. The gating circuit is ready for another operation as soon as charges have returned to normal on the various capacitances in the circuit.

Other common variants of the circuit in Fig. 38 are based on alternative methods of terminating the sweep. For instance, the tap on R_K can be connected to the grid of a pentode which then serves the same role for *T*-3 that *T*-1 does for *T*-2. A more elaborate method (used in the Model 5 synchro-

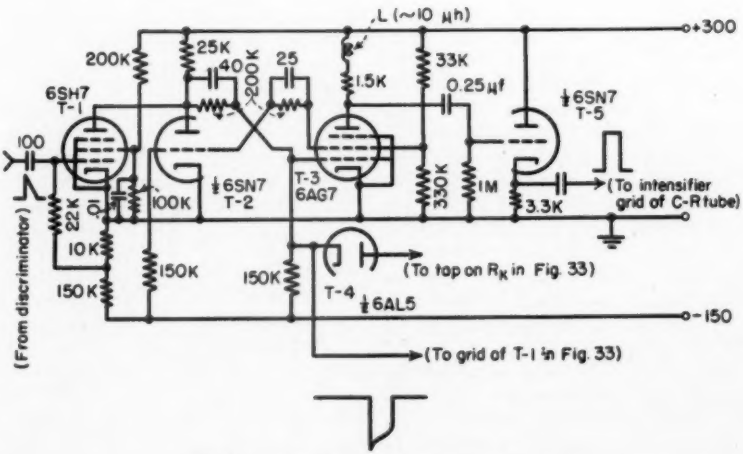

FIG. 38. Gating circuit for sweep generator

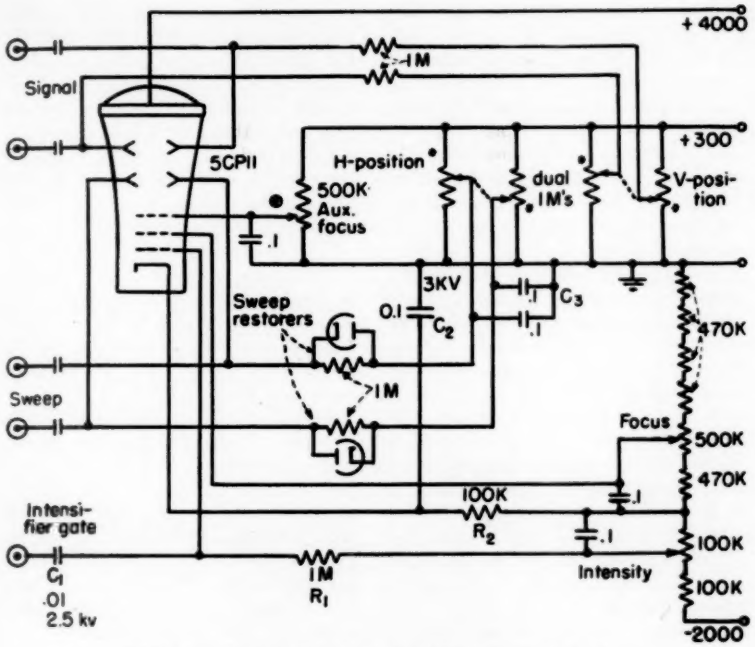

FIG. 39. Typical bias circuit for 5CP11 operated at 6 kv

scope* employs a regenerative amplifier incorporating a blocking oscillator transformer. The signal from the tap on the cathode resistor R_K brings the amplifier grid, via one winding of the transformer, into the conducting region. A fast positive pulse, whose amplitude may be 200 volts or more is coupled from the third winding of the transformer, through a cathode follower, to the grid of *T*-3, thereby quickly terminating the sweep. The second transformer winding, of course, is regeneratively connected in the plate circuit of the amplifier. Although the latter method is very effective in terminating the sweep rapidly, it requires more components than the one shown in Fig. 38. The diode in Fig. 38, incidentally, can be the other half of the diode used in the feedback sweep circuit, Fig. 36, provided the diode heaters are at +300 v (by using the heater supply for the series tube in the voltage stabilizer circuit, for instance).

In a complete sweep circuit based on the various circuit elements described, the following controls are usually provided.

1. Switch for selecting sign of input trigger
2. Discriminator bias potentiometer
3. Sweep speed switch
4. Horizontal centering control
5. Auxiliary focus (see next section) control
6. Length of sweep control (optional)

* Model P5 Synchroscope, built to MIT Radiation Laboratory specifications by the Philharmonic Radio Corporation, New York.

7. Amplitude control for intensifier gate (optional)

In a complete oscillograph, focus and intensity controls, vertical centering, etc., for the C-R tube must of course be provided in addition to the controls most naturally associated with the sweep circuit. Often it is convenient to include a trigger generator with the sweep circuit, for synchroscope style of operation.

2. Bias Circuit for a Cathode-ray Tube. There are several points worth making regarding the design of a bias circuit for a cathode-ray tube. These will be illustrated using the circuit diagram in Fig. 39.

1. The centering voltages are obtained from dual 1 meg potentiometers operated from any convenient 300-v supply used with associated circuits.

2. Anode No. 2 can be adjusted (aux. focus) to make its average potential about equal to that of the deflection plates. A small change in this potential introduces a weak electrostatic lens which may improve the focus of the C-R tube.

3. Diode restorers are shown for the sweep circuit. Note that the capacitors C_3 are necessary for diode restoration.

4. The time constant R_1C_1 of the intensifier (grid) circuit is matched by the time constant R_2C_2 in the cathode circuit. This device is helpful if there exists some ripple voltage on the −2 kv supply, since this ripple will then be fed equally to grid and to cathode when the intensifier is connected to a low-impedance source (as it always is—a cathode follower). If R_3 is not placed in the circuit, the ripple voltage may intensity-modulate the trace (at 60 cps). The resistor R_2 can be omitted if the supply is free from ripple.

5. In critical applications, it is worthwhile to provide a means of adjusting the average potential of one pair of the deflecting plates, to permit a further correction to the focus of the tube by means of a second weak electrostatic (cylindrical) lens (not shown in Fig. 39).

6. The time constants in the leads to the deflection electrodes are made long enough to pass the longest transients to be observed. (The considerations which apply here have already been discussed in connection with amplifier transient response.)

7. It is often desirable to incorporate a diode restorer for the intensifier grid, in order that the intensity control need not be adjusted for different duty ratios of the sweep. The heater of the diode can be connected to the 6.3-v supply for the C-R tube.

3. Amplifiers for Cathode-ray Oscillographs. It is advantageous to employ a "push-pull" amplifier for driving the vertical deflecting plates of a cathode-ray tube. Not only does this mode of operation result in better focussing of the tube, since the average potential of the plates is kept constant, but it permits greater voltage swings with good linearity of operation.

The figure-of-merit for tubes used at the output of the amplifier (to achieve the greatest possible signal amplitude-rise-time quotient) is clearly I_p/C_{out}, since the maximum signal is necessarily less than $R_{load}I_p$ and the rise time can be no smaller than that determined by the constant $R_{load}C_{out}$. In practice, beam power pentodes (or tetrodes) make the best tubes for this application, especially types 6AG7, 807 and 829-B.

To pass from single-sided to push-pull operation requires the use of a phase-inverting (or phase-splitting) stage. If the amplifier is not to employ feedback for stabilization, the type of inverting stage shown in Fig. 40 is particularly satisfactory. The output stages of modern Dumont oscillographs are similar to this circuit element. Phase inversion occurs via the common cathode resistor R_K whose value should

be large compared with the value of $1/g_m$, where g_m is the transconductance of the tubes (perhaps $R_K \approx 10/g_m$).

The circuit element of Fig. 40 is not stabilized by feedback, but can be used over a very wide frequency range, provided proper attention is given to the plate load impedances which are often shunt-compensated. Its gain is slightly less than the gain that either tube would have if R_K were by-passed.

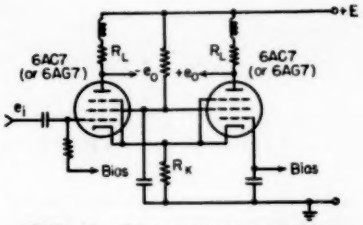

FIG. 40. Phase-splitting amplifier

Where stability and linearity of operation are important, inverse feedback can be used with the output stages. A basic type of circuit which has proved useful is shown in Fig. 41, where tube *T*-1 serves to produce a balanced (push-pull) signal to drive two identical feedback loops, one of which is shown. The gain of the loop is somewhat less than $(R_F + R_K)/R_K$. It will be noticed that a blocking capacitor is not used in the feedback path, to avoid the introduction of a large parasitic capacitance to ground. The omission of this capacitor somewhat complicates the design of such an amplifier as regards d-c currents and voltages, but the design usually can be carried out satisfactorily. The trimmer capacitors shown are used to align the amplifier for good transient response to a step signal.

4. Delay Lines. The present discussion of delay lines will be concerned mostly with an idealized case. It will serve to bring out the essential concepts involved, but will be of little use in the practical design of high-fidelity delay lines, such design usually proceeding in a semi-empirical manner.

Consider the four terminal network

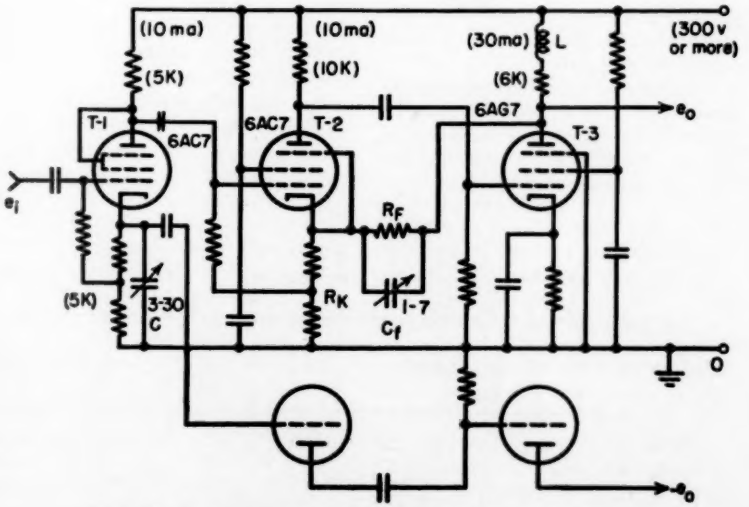

FIG. 41. Output amplifier circuit for a cathode-ray oscillograph

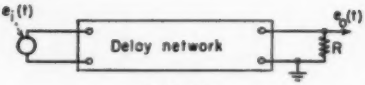

FIG. 42

shown in Fig. 42 where it is desired that

$$e_o(t) = e_i(t - \tau) \qquad (76)$$

and where we can assume that $e_i(t) = 0$ for $t < 0$.

On taking the Laplace transform of Eq. (76) we have that

$$E_o(s) = \int_0^\infty e^{-st} e_i(t - \tau) dt = \int_0^\infty e^{-s(\tau+\lambda)} e_i(\lambda) d\lambda,$$

or

$$E_o(s) = e^{-s\tau} E_i(s). \qquad (77)$$

Hence an ideal delay circuit has the transfer function $e^{-s\tau}$ (with the possible addition of a constant numerical factor to take attenuation into account).

From the point of view of the sinusoidal response of the ideal network, the transfer function becomes (letting $s = j\omega$) $e^{-j\omega\tau}$, so if the input signal is $e^{j\omega t}$, the output signal has the form $e^{j\omega(t-\tau)}$. Consequently the phase shift (lag) of the network is a linear function of the frequency, whereas the amplitude is independent of frequency.

We shall now show that the transfer function $e^{-s\tau}$ can be achieved with the lossless transmission line illustrated in Fig. 43, where L_1 and C_1 are the distributed inductance and the distributed capacitance, respectively, per unit length of a lossless transmission line connecting a source of signals with a terminating resistance R set equal to the characteristic impedance of the lossless line, $(L_1/C_1)^{1/2}$. The network equations for a differential element dx of the line are

$$\left.\begin{aligned} \frac{\partial e}{\partial x} &= -L_1 \frac{\partial i}{\partial t} \\ C_1 e &= -\int \frac{\partial i}{\partial x} dt \end{aligned}\right\} \qquad (78)$$

which give the wave equation for a voltage signal

$$\frac{\partial^2 e}{\partial t^2} = \frac{1}{L_1 C_1} \frac{\partial^2 e}{\partial x^2} \qquad (79)$$

showing that the velocity of propagation is $v = 1/\sqrt{L_1 C_1}$, or that the delay per unit length is $\sqrt{L_1 C_1}$. The Laplace transform of Eq. (79) is

$$\frac{d^2 E}{dx^2} - \frac{s^2}{v^2} E = 0 \qquad (80)$$

which has the general solution

$$E(x,s) = Ae^{\frac{x}{v}s} + Be^{-\frac{x}{v}s}. \qquad (81)$$

where A and B are functions of s (but not of x, of course). From the Laplace transform of the first of Eqs. (78) we find that the current transform corresponding to Eq. (81) is

$$I(x,s) = -\sqrt{\frac{C_1}{L_1}} \left(Ae^{\frac{x}{v}s} - Be^{-\frac{x}{v}s}\right) \qquad (82)$$

Now at the far end, $x = L$, we have

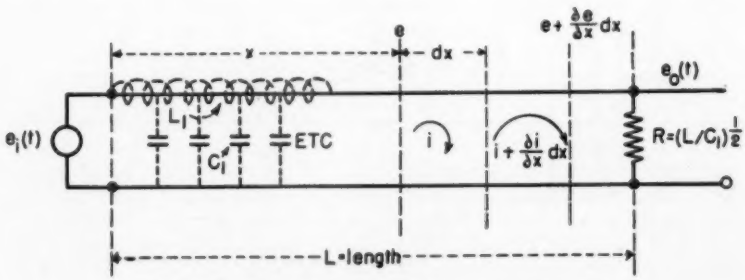

FIG. 43. Circuit for analysis of a transmission line

made

$$\frac{E(L,s)}{I(L,s)} = R = \sqrt{\frac{L_1}{C_1}} = \frac{Ae^{\tau s} + Be^{-\tau s}}{-\sqrt{\frac{C_1}{L_1}}(Ae^{\tau s} - Be^{-\tau s})} \quad (83)$$

where $L/v = \tau$, the total delay. Evidently Eq. (83) requires that $A = 0$, *i.e., terminating the line by its characteristic impedance* $(L_1/C_1)^{1/2}$ *prevents a reflected wave* $\left[A \exp\left(\frac{x}{v}s\right)\right]$ *from travelling back towards the source.* Now at $x = 0$,

$$E(0,s) = B = E_i(s), \quad (84)$$

the Laplace transform of the input voltage. Hence the transform of the output signal is

$$E_o(s) = E(L,s) = Be^{-\tau s} = E_i(s)e^{-\tau s}, \quad (85)$$

which is simply Eq. (77), *i.e.*, an arbitrary input signal is related to the output signal by Eq. (76).

The impedance looking into the terminated line towards the output end at any point between $x = 0$ and $x = L$ is

$$\frac{E(x,s)}{I(x,s)} = \frac{Be^{-\frac{x}{v}s}}{\sqrt{\frac{C_1}{L_1}}Be^{-\frac{x}{v}s}} = \sqrt{\frac{L_1}{C_1}}$$

The above treatment suggests that an ordinary coaxial cable can be successfully used as a delay line. As a matter of fact, for very short delays it is a particularly satisfactory form of delay line, but for delays longer than a few tenths of a microsecond an unduly long cable is required.

The obvious way of making a compact delay line is to increase the inductance and capacitance per unit length, for instance, by mounting a number of strips of metal foil along the inside or outside of a long slender solenoid, as suggested in Fig. 44. The distributed capacitance of these strips to a unit length of the winding equals C_1. The capacitive electrode is subdivided to

FIG. 44. Manner of constructing a delay line

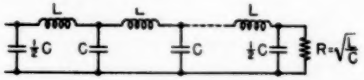

FIG. 45. Lumped-parameter delay line

prevent a short-circuited turn from surrounding the solenoid. Still another way of making a delay line consists of separating the inductance and capacitance as indicated in Fig. 45, where the delay per section is $\sqrt{LC}$ and the (nominal) characteristic impedance is $\sqrt{L/C}$. The resulting lumped parameter delay line is used mostly for delaying trigger pulses, since it distorts a fast pulse by superposing transient oscillations on it.

An elaborate corrective network is sometimes employed in order to improve the characteristics of a delay line. Even so a step function signal cannot be propagated along the line without its steep front gradually spreading out. It would appear that part of the difficulty, at least, lies in mutual inductance between adjacent parts of the line (if it is of the continuous sort). One would then expect the steep front to spread out until its length along the line has the order of magnitude of the diameter of the solenoid. All practical delay lines deviate from the ideal line, as shown by a finite rise time for a step function input signal. A high quality line, however, will possess very nearly a monotonic transient response. The figure-of-merit of a delay line is defined by the delay-time-rise-time quotient. Values of this quotient in the range 10 to 50 are common, *i.e.*, a delay of 2 μsec

with a rise time somewhat less than 0.1 μsec is possible.

It is desirable to drive a delay line from a low-impedance source such as a cathode follower, and at a signal level of not more than a few volts because of the low characteristic impedance of the line. If difficulty is experienced with the reflection of small, high-frequency components of a fast signal from the terminated end, some improvement usually results by insuring that the input end is driven from a source having a resistance equal to the characteristic impedance of the line. A loss in signal amplitude results if the input end, as well as the output end, is properly matched, unless the source happens to have the correct impedance, without the addition of a series or parallel resistance.

A common method of driving a delay line is simply to make it the plate load of a pentode amplifier stage. In this case the correct termination of the input end requires a parallel resistance equal to $(L_1/C_1)^{1/2}$ (since the plate resistance of the tube is so extremely high). This method does not make use of the feedback stabilization present in the cathode follower, and should not be used when the rest of the amplifying system is of the feedback type. Even in the case of the cathode follower, linearity and stability are sacrificed when the cathode load is made comparable with $1/g_m$. For this reason it is sometimes desirable to drive a delay line from the output of one of the feedback amplifier loops described earlier.

Commercial delay lines have been designed mostly for radar applications. Two of the most useful lines are the Sickles No. 12647 and the Western Electric No. D-163169. The first of these lines has a delay of 1.2 μsec, and a rise time of about 0.06 μsec, and the second, a delay of 4.5 μsec, and a rise time of about 0.1 μsec. Their characteristic impedences are 350 and 650 ohms, respectively.

In conclusion it is worth remarking that band widths quoted by manufacturers of delay lines often give an erroneous impression of the rise time of the line, *i.e.*, the result $T_R \approx \frac{1}{3\Delta f}$ cannot be applied to this information.

XI. Delay-line Pulse Shaping

It has already been pointed out that an amplifier used with a detector of radiation is designed to convert a series of voltage steps into a series of discrete pulses suitable for sorting as to amplitude, frequency of occurrence, distribution in time, etc. The common method of shaping pulses consists in decreasing *one* of the RC time constants in the amplifier to a point where an input step signal emerges as a pulse as short as is necessary. Such a pulse, however, does not have an ideal shape, on account of the long duration of the trailing edge in comparison with the short duration of the top of the pulse. In certain applications it is necessary to have pulses approximating a rectangle in shape, indeed a rectangular-shaped pulse may be considered to have the best possible form for nearly all counting applications.

The basic idea involved in the use of a delay line to shape pulses can be described as follows. At some point in the amplifier the signal is fed into two channels. One channel contains a delay line of delay T seconds, and has an amplification $+A$. The other channel has an amplification $-A$ and no delay line. Provided the input signal to the amplifier consists of a step function, and the rise time of the amplifier is much shorter than the time T, then the signal produced by adding the output signals from the two channels evidently con-

sists of a rectangular-shaped pulse lasting a time T. In practice it is found that this result can be achieved more simply by employing the delay line shorted at one end as a coupling impedance in the amplifier.

To explain this alternative method, let us assume for the time being that the delay line is an ideal line, *i.e.*, that it is loss-less and does not distort a step signal. We next suppose that the line is connected in a circuit having the equivalent circuit shown in Fig. 46, where e_i is the step-signal input, e_0 is the output pulse, and $R = Z_c$ is the characteristic impedance of the delay line. At the onset of a step signal, the delay line presents its characteristic impedance Z_c (a pure resistance), so $e_0 = \frac{1}{2} e_i$. Simultaneously a wave starts down the line and is reflected with change of phase from the shorted end. At a time $2T$ the reflected wave arrives at the input end where it is completely absorbed by the series terminating resistance R. The delay line now acts as a short circuit and the output signal drops to zero. The step signal evidently has been converted into a rectangular-shaped pulse lasting $2T$.

This somewhat unconventional way of viewing the behavior of the shorted delay line (*i.e.*, as a resistance which has first one value, and then another) is useful in considering the practical case in which the delay line has a finite d-c resistance R_0. In such event, it is clear that the output signal will not fall to zero at time $2T$, but to the value $(R_0/R_0 + Z_c)e_i$. The deviation from ideal behavior must be corrected for if the method is to be of much use. It turns out that either of two devices can be adopted to remedy the defect. (1) The shorted delay line can be made one arm of an impedance bridge which is balanced for slowly changing signals. (2) The step signal can be "differentiated" slightly with an RC circuit so that the signal falls off exponentially

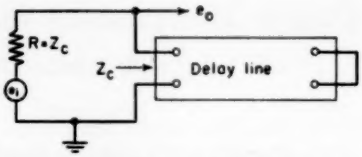

FIG. 46. Equivalent circuit for delay-line pulse shaping

at such a rate that its amplitude has decreased just enough in the time interval $2T$ to equal the reduced amplitude of the reflected wave. The cancellation is again made perfect, but the top of the formerly rectangular-shaped pulse now has a slight negative slope. Method (2) is the one usually adopted for compensating for the d-c resistance of the delay line.

The finite rise time of the delay line contributes to the over-all rise time of the amplifying system in the manner discussed in an earlier section, and requires no additional comment here. High quality delay lines designed for delaying transients can be used for pulse shaping, or special lines can be constructed. In selecting a suitable line, the ratio Z_c/R_0 should be chosen to be as large as possible to reduce the amount of compensation needed. Delay-line pulse shaping is best applied when it is desired to produce pulses whose duration is not more than perhaps 10 μsec. A study of the signal-to-noise ratio of pulse amplifiers by the author indicates that this ratio is highest when the delay $2T$ is approximately equal to the rise time of the amplifier. A pulse then has a shape approximating that of a Gauss error curve, and not that of a rectangle.

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Dosage Determination with Radioactive Isotopes—I*

FUNDAMENTAL DOSAGE FORMULAE

When radioactive isotopes are employed either as tracers or in therapy, it is important to be able to determine the radiation dosage. This cannot, in general, be measured, but when the half-life, radiation energy, and biological uptake and excretion are known, it can be calculated.

The paper is divided into two parts,† based on physical and clinical aspects of the problem. In the physical part (Part I) are given mathematical developments of formulae for dosage rates and total doses for beta- and gamma-ray-emitting isotopes, together with subsidiary formulae for safe concentration. In the clinical part (Part II) these formulae are applied for dosage considerations for specific cases, with isotopes of common interest. Particular consideration is given to the determination of safe tracer doses.

Since the basic information regarding radiation disintegration schemes and energies is scattered through many journals, it has been considered desirable to collect pertinent data. Two extensive tables are presented, for beta and gamma rays, respectively, giving half-life, average radiation energy, fraction disintegrating per day, and specific dosage data, including the safe tracer concentration, for some 38 isotopes.

A KNOWLEDGE of radiation dosage from artificially radioactive substances deposited in tissue is important in applications of these materials to tracer studies, as well as in therapy. In employing tracers in humans, it is essential to know that the incidental irradiation does not exceed safe limits. In all tracer work it must be assured that the radiation itself does not affect the phenomenon being investigated. While this can actually be determined only by trial, a knowledge of the tissue doses involved is useful. In therapy the same considerations apply as in any other type of irradiation.

It is highly desirable to express these doses in terms of roentgens, since that unit is generally employed for therapy with X-rays and radium. In the case of X-rays, ionization measurements in phantoms give satisfactory dosage data. For radium and radon in sealed containers, extensive dosage tables have been calculated, based on an experimentally determined value of radiation from a standard point source. Similar methods would be applicable to any gamma-ray emitting radioactive substance enclosed in a sealed container and used in the same manner as radon.

As commonly used, most radioisotopes are ingested or injected in soluble form and subsequently deposited with a greater or lesser degree of selectivity in various cells or organs. It is evident that neither measurements nor calculations of the types cited would be satisfactory under such conditions.

* This paper is based in part on work done under contract N6-ORI-99 with the Office of Naval Research.

† Part II will be published next month.

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Nevertheless when the physical factors of half-life and radiation energy, and the physiological factors of uptake and excretion are known, it is possible, in some cases at least, to make satisfactory estimates of tissue dosage.

Beta-ray and Positron Emitters

The *roentgen*, as defined by international agreement,‖ applies only to X- or gamma radiation; it can therefore be used for gamma-ray emitting isotopes but not for radiation due to primary beta particles. On the other hand, if the energy absorbed per gram of air per roentgen (~83 ergs) is made the unit of energy absorption for beta rays, it is possible to establish a comparable basis for beta-ray dosage. To be sure, in going from air to tissues certain corrections will have to be made because the energy absorption in tissue per roentgen exposure of X- and gamma rays depends on both tissue composition and radiation wavelength, but in practice, for soft tissues, these corrections are not very large. Hence, it is possible to define an "equivalent roentgen" as "that amount of beta radiation which, under equilibrium conditions, releases in one gram of air as much energy as one roentgen of gamma rays." Since the accepted symbol for the roentgen is "r," it is convenient to designate the equivalent roentgen by "e.r." (This is essentially the same unit as the "rep" or "roentgen equivalent physical" of the Plutonium Project.)

Consider a mass of tissue, of linear dimensions large as compared to the range of ordinary beta particles, in which a radioelement is concentrated uniformly and is not being eliminated. Except for regions close to the boundary of the mass, the energy D_β absorbed per gram of tissue will be

$$D_\beta = n\bar{E}_\beta \times 10^6 \text{ ev} \qquad (1)$$

where

$n = C/\lambda$ is the number of radioactive atoms per unit mass and

$\bar{E}_\beta$ is the *average* beta-ray energy per disintegration in Mev.

$n = 3.7 \times 10^4 \times C/\lambda$ if C is the concentration in μc¶ per gram (or mc/kg) and λ the decay constant of the element in sec^{-1}.

The energy E_r absorbed per gram of *air* when the latter is exposed to one roentgen of radiation is

$$E_r = N_a \times W = 1.62 \times 10^{12} \times 32.2 = 5.22 \times 10^{13} \text{ ev} \qquad (2)$$

where

$N_a = 1.62 \times 10^{12}$, the number of ion pairs formed in a gram of air per roentgen and

$W = 32.2$ ev, the energy required to produce one pair of ions in air.

It follows from (1) and (2) that D_β can be expressed in equivalent roentgens by simple division, namely

$$D_\beta = \frac{3.7 \times 10^{10} C\,\bar{E}_\beta}{5.22 \times 10^{13} \times \lambda} \text{ e.r.} \qquad (3)$$

This expression can be modified into one more easily remembered if the half-life of the isotope is expressed in days, as

$$T = \frac{0.693}{\lambda \times 8.64 \times 10^4}$$

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‖ The roentgen is defined as "that quantity of X- or gamma radiation such that the associated corpuscular emission per 0.001293 gram of air produces in air ions carrying 1 esu of quantity of electricity of either sign."

¶ A microcurie of any radioactive element is understood to be that quantity that disintegrates at the rate of 3.7×10^4 atoms/sec.

Equation 3 now becomes

$$D_\beta = 88\overline{E}_\beta TC = K_\beta C \text{ e.r.} \quad (4)$$

where

$K_\beta = 88\ \overline{E}_\beta T$ e.r. per μcd*

per gram of tissue (4a)

K_β gives immediately the dose in equivalent roentgens for each microcurie completely disintegrated within a gram of tissue.

From this formula others can be easily derived. For any interval of time t the dose d_β is proportional to that fraction of atoms disintegrated during that time, namely

$$d_\beta(t) = D_\beta(1 - e^{-\frac{0.693\,t}{T}}) \text{ e.r.} \quad (5)$$

For $T > 5$ days, equation 5 can be written with an approximation of better than one percent as

$$d_\beta = 61\ \overline{E}_\beta C \text{ e.r. per day} \quad (6)$$

Similarly for $T > 5$ hours

$$d_\beta = 2.54\ \overline{E}_\beta C \text{ e.r. per hour} \quad (7)$$

Equation 5 is particularly useful in computation of the concentration necessary to deliver the so-called *tolerance dose of* 0.1 *r per day*. Thus, if S_β is defined as the concentration in microcuries per kg of tissue weight necessary to deliver the tolerance dose in the first day, it follows that

$$S_\beta = \frac{0.1 \times 10^3}{K_\beta \times f_d} \quad \mu\text{c per kg of tissue weight} \quad (8)$$

In this expression f_d is the fraction of atoms disintegrating in one day, namely:

$$f_d = (1 - e^{-\frac{0.693}{T}}) \quad (9)$$

Data regarding beta-ray emitters are presented in Table 1. Z and A are the atomic numbers and weights, respectively. $\overline{E}_\beta$, K_β and f_d have the meanings stated above. The last column gives the ranges in water for the most energetic beta particle of each beta-ray spectrum for the different isotopes.

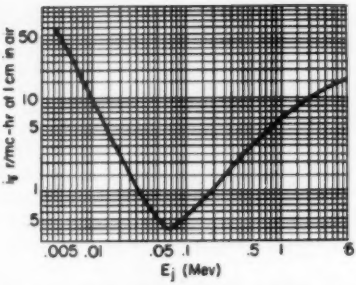

FIG. 1. i_γ in roentgens per mc-hour at 1 cm distance in air as a function of the gamma-ray energy E_j in Mev

Attention should be directed to column 7 which indicates the safe tracer concentration of 0.1 r for the first day, due to the beta rays alone. The gamma-ray contribution will be discussed below. In the case of a short-lived element, the dose on succeeding days will quickly decrease to the vanishing point, while for long-lived substances it will continue at an appreciable level for some time.

The values of $\overline{E}_\beta$ were calculated as outlined in a previous paper by two of the present authors (*1*).† It is to be realized that $\overline{E}_\beta$ depends on the disintegration scheme of the isotope under consideration and that the latter is often complex. The disintegration schemes of some isotopes are established with satisfactory accuracy and these isotopes constitute Group A of Table 1; those isotopes for which the schemes are not yet well known comprise Group B. Of these, Sr^{90} is of particular interest since it decays into Y^{90} which is also a radioactive element (*2*). The

* Microcuries destroyed, *i.e.*, microcuries remaining in the tissue until completely disintegrated.

† In this paper (*1*) references are given for the disintegration schemes of a large number of isotopes. A supplementary list of references, for the additional isotopes considered in Tables 1 and 2, is appended to the bibliography of the present paper.

given values of $\overline{E}_\beta$ in Group C of Table 1 include the total localized X-radiation following the decay by electron capture. The X-radiation of the elements in this group is so soft that the resulting ionization is limited to a few mm of tissue. $\overline{E}_\beta$ in Group D consists of that part of the radiation released by electron capture, which is available only as Auger electrons and soft *L*-radiation.‡

Gamma-Ray Emitters

The determination of the dosage due to gamma radiation, originating from a stable distribution of a radioelement in tissue, is greatly facilitated by the calculation of the constant I_γ which expresses the *exposure in roentgens at 1 cm distance from an unfiltered unit point source in air during unit time interval.* I_γ can be determined by calculating the ionization produced by a point source in one cubic centimeter of air.

In free air, if a spherical shell of 1 cm radius and thickness dr cm has at its center a point source of 1 mc of an isotope emitting a gamma ray of energy E_j in a fraction P_j of its disintegrations, that shell will receive energy at the rate

$$dF_j = 3.7 \times 10^7 P_j N_e E_j \mu_j^e \, dr \quad \text{(ev/sec)} \quad (10)$$

where

$N_e = 3.9 \times 10^{10}$ electrons per cc of air

$$E_j\mu_j^e = E_j(\sigma_a + \tau)_j + (E_j - 2m_0c^2)(\sigma_p)_j.$$

μ_j^e is the absorption coefficient per electron§; σ_a, τ and σ_p are the scattering absorption, photoelectric absorption and pair production coefficients per electron, respectively; and m_0c^2 is the rest energy of the electron.

The rate of energy absorption δF_j per cc of air (1.293 mg) under standard conditions is:

$$\delta F_j = \frac{3.7 \times 10^7}{4\pi} \times P_j N_e E_j \mu_j^e \quad \text{(ev/cc/sec)}$$

Absorption of this energy will result in ionization, the measure of which in esu of current, is given by

$$(i_\gamma)_j = \frac{\delta F_j}{n_i \times W} = \frac{3.7 \times 10^7 N_e}{4\pi n_i W} \times P_j E_j \mu_j^e \quad (11)$$

where

$n_i = 2.08 \times 10^9$, the number of ion pairs per esu of current, and

$W = 32.2$ ev, the energy required to produce one pair of ions in air.

From the definition of the roentgen it follows that equation 11 expresses numerically the exposure in roentgens per second at 1 cm distance from a point source of 1 mc emitting P_j gamma rays of energy E_j per disintegration. A more convenient formula results if the hour is chosen as the unit of time and E_j is expressed in Mev. Formula 11 then becomes

$$(i_\gamma)_j = 6.14 \times 10^{25} P_j E_j \mu_j^e \text{ roentgens per mc-hr at 1 cm distance in air} \quad (12)$$

A plot of $(i_\gamma)_j$ as a function of E_j (for $P_j = 1$) is shown in Fig. 1.

If the isotope has a complex spectrum of gamma rays of different energies E_j, each type emitted with probability P_j, then (12) becomes

$$I_\gamma = \sum_j (i_\gamma)_j$$

$$I_\gamma = 6.14 \times 10^{25} \sum_j P_j E_j \mu_j^e \text{ roentgens per mc-hr at 1 cm distance in air} \quad (13)$$

‡ A more detailed discussion of the doses obtained by the elements in Group C and Group D is included in another paper by the present authors (3).

§ The electronic absorption coefficient μ_j^e and the linear absorption coefficient μ_j used in this paper do not include that portion of the total absorption coefficient which relates to the scattered radiation of the Compton process. This procedure is entirely justified for a few cm of water (4) and introduces no appreciable error for at least 8 cm of Al, when the radiation is of hardness comparable to the gamma rays of radium (5).

TABLE 1

Physical data pertaining to calculations of radiation dosage resulting from beta rays and/or very soft X-ray radiation. The values of $K\beta$ and $S\beta$ are based on uniform and biologically stable concentrations of radioelements distributed in tissues of linear dimension large as compared to the range of the beta particles. The sign "0" under the heading "Radiation" indicates the absence of nuclear gamma rays.

$\bar{E}\beta$ is the average energy per disintegration.

$K\beta = 88\bar{E}\beta T$ is the radiation dose expressed in equivalent roentgens due to

Element	*Z*	*A*	*Radiation*	*T Half-life in days*	$\bar{E}\beta$ *(Mev)*
Group A: $\bar{E}\beta$ is known to an accuracy of a few per cent					
C	6	11	β^+, 0	0.014	0.380
N	7	13	β^+, 0	0.007	0.475
Na	11	22	β^+, γ	1100	0.225
		24	β^-, γ	0.61	0.540
P	15	32	β^-, 0	14.5	0.695
Cl	17	38	β^-, γ	0.026	1.390
K	19	42	β^-, γ	0.515	1.395
Sc	21	46	β^-, γ	85	0.117
V	23	48	β^+, K, γ	16	0.175
Mn	25	52	β^+, K, γ	6.5	0.085
		56	β^-, γ	0.108	0.890
Fe	26	59	β^-, γ	47	0.120
Co	27	56	β^+, γ	85	0.655
		60	β^-, γ	1940	0.099
Cu	29	61	β^+, K, 0	0.142	0.433
		64	β^+, β^-, K, 0	0.53	0.120
Zn	30	63	β^+, K, γ	0.027	0.965
As	33	76	β^-, γ	1.12	1.170
Br	35	82	β^-, γ	1.5	0.150
In	49	114	β^-, (γ)	50	0.940
I	53	130	β^-, γ	0.525	0.270
		131	β^-, γ	8.0	0.205
RaE	83	210	β^-, 0	4.85	0.330
Group B: $\bar{E}\beta$ is less accurately known than $\bar{E}\beta$ in Group A					
C	6	14	β^-, 0	$1.8 \cdot 10^6$	0.05
S	16	35	β^-, 0	88	0.055
Ca	20	45	β^-, 0	180	0.085
Sr	38	89	β^-, 0	55	0.57
		90	β^-, 0	9000	0.22
Y	39	90	β^-, 0	2.6	0.90
Sb	51	124	β^-, γ	60	0.66
I	53	128	β^-, γ	0.017	0.77
Au	79	198	β^-, γ	2.7	0.32
Group C: $\bar{E}\beta$ includes the total localized X-radiation following decay by electron					
Mn	25	54	K, γ	310	0.0054
Fe	26	55	K, γ	1500	0.0059
Co	27	58	β^+, K, γ	65	0.035
Zn	30	65	β^+, K, γ	250	0.01
Group D: $\bar{E}\beta$ consists of part of the radiation released in the decay by electron					
Y	39	86	K, γ	105	0.005
In	49	111	K, γ	2.7	0.0058

beta rays emitted during the complete disintegration of 1 μc of radioelement per gram of tissue.

$f_d = \left(1 - e^{-\frac{0.693}{T}}\right)$ is the fraction of the entire quantity of isotope which disintegrates in 24 hours

$S\beta = \frac{0.1 \times 10^3}{K\beta \times f_d}$ is the concentration of radioisotope expressed in μc/kg which will deliver a dose of 0.1 e.r. to tissue during the first 24 hours of exposure.

Kβ e.r./μcd per gram	*f_d fraction disintegr. per day*	*Sβ μc per kg*	*Weight per me in 10^{-9} gram*	*Maximum range in water (mm)*
0.47	1.0	213	0.0012	4.2
0.29	1.0	345	0.0007	5.6
22000	$6.3 \cdot 10^{-4}$	7.3	197	2.1
29	0.68	5.1	0.113	6.4
885	0.047	2.4	3.6	8.0
3.2	1.0	31	0.0076	27
63	0.74	2.1	0.167	19
870	0.008	14.3	30	1.0
245	0.042	9.7	5.9	2.8
48	0.101	20.6	2.6	2.2
8.5	0.998	11.8	0.046	14
496	0.015	13.4	21.3	1.5
4900	0.008	2.6	36.6	7.0
17000	$3.6 \cdot 10^{-4}$	16.5	895	0.8
5.4	0.992	18.7	0.067	5.5
5.6	0.73	24.4	0.26	2.6
2.3	1.0	43.5	0.013	12
115	0.46	1.9	0.655	15.7
20	0.37	13.5	0.95	1.6
4150	0.014	1.7	44	9.4
12.4	0.73	11.0	0.53	4.5
144	0.083	8.3	8.1	2.2
141	0.133	5.3	7.85	5.2
$8 \cdot 10^6$	$4 \cdot 10^{-7}$	32	$18 \cdot 10^4$	0.24
420	0.0079	30	24	0.2
1350	0.0039	19	62	0.6
2760	0.013	3	38	7
$17 \cdot 10^4$	$8 \cdot 10^{-5}$	8	6200	2.2
200	0.24	2	18	11
3480	0.012	2.4	57	12.3
1.2	1.0	87	0.017	9.8
76	0.23	5.7	4.1	3.8
capture				
147	0.0022	340	128	
780	$4.6 \cdot 10^{-4}$	280	633	
20	0.012	415	29	1.5
180	0.003	185	124	1.2
capture				
46	0.007	310	69	
1.4	0.23	310	2.3	0.01

The knowledge of the constant I_γ permits the prompt measurement in millicuries of some gamma-ray emitting isotopes, by the simple measurement of the dosage rate in roentgens. This constant is furthermore indispensable for the knowledge of therapeutic doses administered by discrete sources of known millicurie content.*

With the knowledge of I_γ the problem of tissue dosage determination is not yet solved. When the radioelement, by virtue of ingestion, injection, etc., is distributed throughout a tissue volume V with a uniform and biologically stable concentration of c millicuries per gram, the total dose rate per hour at any point 0 due to the isotope in the whole tissue is

$$d_\gamma = \sum_j c(i_\gamma)_j \rho \int_V \frac{e^{-\mu_j s}}{s^2}\, dV$$

roentgens per mc-hr (14)

s is the distance of the volume element dV from point 0, ρ is the density of the tissue, and μ_j the linear absorption coefficient.†

The values of the linear absorption coefficient μ_j for water are plotted in Fig. 2 as a function of the gamma-ray energy E_j.

The integral in equation 14 will be called the *geometrical factor* g_j:

$$g_j = \rho \int_V \frac{e^{-\mu_j s}}{s^2}\, dV$$

With the values of $(i_\gamma)_j$ and μ_j given in Figs. 1 and 2, the total dose rate d_γ can now be calculated for each gamma-ray energy E_j.

In the energy range of most of the gamma rays given in Table 2, *e.g.*, between 0.08 and 2 Mev, the absorption

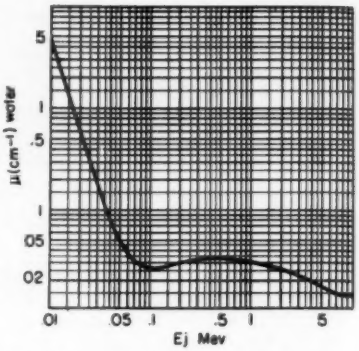

FIG. 2. Linear gamma-ray absorption coefficient μ_j in water as a function of the gamma-ray energy E_j. (μ_j includes only the absorption caused by the liberation of photoelectrons, Compton electrons and electron pairs, but not that fraction due to Compton scattering)

coefficient μ_j is fairly constant ($\mu_j \sim 0.03$, see Fig. 2). For therapeutic dosage and protection estimates, equation 14 can therefore be written as

$$d_\gamma = I_\gamma c g \text{ roentgens per mc-hour} \quad (15)$$

Finally the total dose evaluated per microcurie destroyed (μcd) for a uniform and biologically stable concentration of C microcuries per gram of tissue follows from equation 15 as

$$\underline{D_\gamma = 1.44 t I_\gamma \times 10^{-3} C g = K_\gamma C g}$$

roentgens per C μcd per gram of tissue (16)

in which t is the half-life of the isotope in hours, and

$$\underline{K_\gamma = 1.44 t I_\gamma \times 10^{-3}} \text{ roentgens per } \mu\text{cd at 1 cm in air} \quad (16a)$$

The values of I_γ, K_γ and other pertinent data for gamma-ray emitters are presented in Table 2. In this table, the dose due to the annihilation radiation (two gamma rays of 0.511 Mev each per positron emitted) resulting from positron-electron recombination is included in the values given for I_γ.

The *geometrical factor*, g, must be calculated for each specified point; for

* It should be kept in mind that a positron emitter is always a gamma-ray source, because of the annihilation radiation, even if there is no nuclear gamma radiation.

† See second footnote on page 59.

the sake of illustration g will be evaluated for two simple cases. (The density of tissue is assumed to be unity.)

1. 'g' at the midpoint on the axis of a cylinder. The integral for the geometrical factor g_j at the midpoint on the axis of a cylinder of radius R and height $2Z$ is

$$g_j = 4\pi \int_0^Z \int_0^R \frac{r dr dz}{r^2 + z^2} e^{-\mu_j \sqrt{r^2 + z^2}} \tag{17}$$

For small values of $\mu_j \sqrt{R^2 + Z^2}$ as a satisfactory approximation

$$g_j = 2\pi \left\{ Z \ln \left(1 + \frac{R^2}{Z^2}\right) + 2R \tan^{-1}\left(\frac{Z}{R}\right) - \mu_j \left[Z \sqrt{R^2 + Z^2} - Z^2 + R^2 (\ln [Z + \sqrt{R^2 + Z^2}] - \ln R) \right] \right\} \tag{18}$$

As an example, for $Z = 30$ cm and $R = 20$ cm, equation 18 becomes

$$g_j = 314 - 4140\mu_j$$

2. 'g' at different points inside and outside of a sphere. For the simplest geometrical shape, namely a sphere, it is possible to present the geometrical factor graphically for different points inside and outside of the sphere as a function of its radius. The given curves furnish a good approximation for this constant for a considerable number of radioisotopes.

For the *center* of a sphere of radius R

$$g_j = \int_0^R \frac{4\pi s^2 ds}{s^2} e^{-\mu_j s} = \frac{4\pi}{\mu_j} (1 - e^{-\mu_j R}) \tag{19}$$

For a sphere of radius $R < 10$ cm, an approximate value of g_j at the center, correct to within a few per cent is:

$$g_j = 4\pi R \left(1 - \mu_j \frac{R}{2}\right) \sim 4\pi R \tag{20}$$

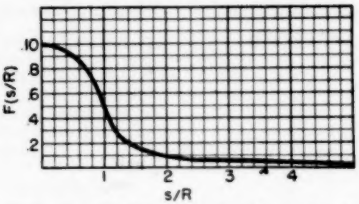

FIG. 3. F(s/R) as a function of s/R (Equation 22)

The geometrical factor can be calculated to a first approximation for any *point at a distance* s *from the center of the sphere* (*6*, *7*).*

For $s \leq R$

$$g_j = 2\pi R \left\{ \left[1 + \frac{R^2 - s^2}{2sR} \ln \frac{R + s}{R - s} \right] - \mu_j R \right\} \tag{21}$$

For $s = 0$, *e.g.*, for the center of the sphere, equation 21 becomes obviously identical with equation 20.

For $s = R$, $g_j = 2\pi R(1 - \mu_j R)$. Hence, neglecting the absorption term $\mu_j R$, the radiation dose at any point on the surface of the sphere is one half of the value at its center when the gamma-ray emitting material is uniformly distributed throughout the volume.

For $s > R$ the geometrical factor is also given by equation 21 with the logarithmic term replaced by $\ln \frac{R + s}{s - R}$, if it is assumed that, outside the sphere, the absorption of the radiation can be neglected.

The absorption term $(\mu_j R)$ in equation 21 is constant throughout any particular sphere. Therefore equation 21 can be written as

$$g_j = 4\pi R \left\{ F\left(\frac{s}{R}\right) - \frac{R}{2} \mu_j \right\} \tag{22}$$

* Many calculations of the geometrical factor can be found in the radium therapy literature. See for example ref. 7.

TABLE 2

Physical data pertaining to calculations of radiation dosage resulting from gamma rays. The sign "0" under the heading "Radiation" indicates the absence of nuclear gamma rays. The column headed "Annihilation Radiation" refers to positron-electron recombination; the column headed "Nuclear Gamma Radiation" refers to gamma rays originating in the disintegrated nucleus. The numbers in parentheses indicate the number of photons of the particular energy that are released per disintegration. In Group B, the numbers in square brackets pertain to X-ray emission following electron capture.

Element	*Z*	*A*	*Radiation*	*t Half-life in hours*	*E_i in Mev Annihilation Radiation*
Group A: elements not decaying by electron capture, or X-ray emission following					
C	6	11	β^+, 0	0.33	0.511(2)
N	7	13	β^+, 0	0.17	0.511(2)
Na	11	22	β^+, γ	26500	0.511(2)
		24	β^-, γ	14.7	
Cl	17	38	β^-, γ	0.62	
K	19	42	β^-, γ	12.4	
Sc	21	46	β^-, γ	2040	
V	23	48	β^+, K, γ	384	0.511(1.16)
Mn	25	52	β^+, K, γ	156	0.511(0.7)
		56	β^-, γ	2.59	
Fe	26	59	β^-, γ	1128	
Co	27	56	β^+, γ	2040	0.511(2)
		60	β^-, γ	46500	
Cu	29	61	β^+, K, 0	3.4	0.511(1.56)
		64	β^+, β^-, K, 0	12.8	0.511(0.38)
Zn	30	63	β^+, K, γ	0.64	0.511(1.86)
As	33	76	β^-, γ	26.8	
Br	35	82	β^-, γ	36	
Sb	51	124	β^-, γ	1440	
I	53	128	β^-, γ	0.42	
		130	β^-, γ	12.6	
		131	β^-, γ	192	
Au	79	198	β^-, γ	65	
Group B: elements with X-ray emission following electron capture whose contri-					
Mn	25	54	K, γ	7450	
Fe	26	55	K, γ	36000	
Co	27	58	β^+, K, γ	1560	0.511(0.3)
Zn	30	65	β^+, K, γ	6000	0.511(0.03)
Y	39	86	K, γ	2530	
In	49	111	K, γ	65	

The function $F(s/R)$ is plotted in Fig. 3; it represents g_i at any point s/R from the center of the radioactive sphere as a fraction of its value at the center, without the absorption term.

The following example will illustrate the application of equation 22 and Fig. 3:

gamma-ray energy,

$$E_i = 0.511 \text{ Mev}$$

absorption coefficient,

$$\mu_i = 0.033 \text{ cm}^{-1} \text{ (Fig. 2)}$$

I_γ is the exposure in roentgens at 1 cm distance in air from an unfiltered point source of 1 mc, for one hour; or milliroentgens per microcurie-hour (see formula 13).

$K_\gamma = 1.44 t I_\gamma \times 10^{-3}$ is the number of roentgens at 1 cm distance in air from an unfiltered point source, per microcurie destroyed.

$f_d = \left(1 - e^{-\frac{0.693}{T}}\right)$ is the fraction of the entire quantity of isotope which disintegrates in 24 hours. (T is the half-life in days)

E_j in Mev Nuclear Gamma Radiation				I_γ at 1 cm mr/μc-hr r/mc-hr	K_γ at 1 cm r/μcd	f_d fraction disintgr. per day
electron capture so soft that it can be treated like beta radiation and hence making no significant contribution to I_γ						
				6.2	0.003	1.0
				6.2	0.0015	1.0
1.30(1)				13.2	500	$6.3 \cdot 10^{-4}$
1.38(1)	2.76(1)			19.1	0.40	0.68
1.6(0.36)	2.15(0.47)			7.6	0.0068	1.0
1.51(0.25)				1.95	0.035	0.74
0.90(1)	1.12(1)			11.4	33.5	0.008
0.98(1)	1.33(1)			16.3	9.0	0.042
0.736(1)	0.94(1)	1.46(1)		19.5	4.4	0.101
1.77(0.3)	2.06(0.2)			9.4	0.035	0.998
1.1(0.5)	1.3(0.5)			6.55	10.7	0.015
0.845(1)	1.26(1)			17.95	37.2	0.008
1.16(1)	1.32(1)			13.5	900	$3.6 \cdot 10^{-4}$
				4.8	0.024	0.992
				1.2	0.022	0.73
0.96(0.07)	1.90(0.01)			6.3	0.006	1.0
0.55(0.37)	1.20(0.12)	1.75(0.01)		2.2	0.083	0.46
0.547(1)	0.787(1)	1.35(1)		15.1	0.79	0.37
0.6(1)	1.7(0.5)			7.9	16.4	0.012
0.428(0.07)				0.2	0.00012	1.0
0.416(0.55)	0.537(1)	0.667(1)	0.744(1)	13.05	0.237	0.73
0.080(1)	0.367(1)			2.65	0.735	0.083
0.40(1)				2.4	0.22	0.23
bution to I_γ (given in square brackets) is not negligible						
0.835(1)		[0.0054(1)]		4.9+[11]	52	0.0022
0.07(2·10⁻⁵)		[0.0059(1)]		[10]		$4.6 \cdot 10^{-4}$
0.805(1)		[0.0064(0.85)]		5.7+[7]	12.8	0.012
1.14(0.46)		[0.008(0.99)]		3.0+[5]	26	0.003
0.908(1)	1.89(1)	[0.0142(1)]		14.4+[3.1]	52.5+[10.3]	0.007
0.173(1)	0.247(1)	[0.0231(1)]		2.3+[1.4]	0.22+[0.13]	0.23

radius of the sphere,

$R = 2$ cm

distance from the center,

$s = 1$ cm

$F(s/R) = 0.91$ (Fig. 3)

$g_j = 4\pi \times 2(0.91 - 0.033 \times 1) = 22.1$

It is now possible to calculate from equations 16 and 22 the gamma-ray dosage per μcd/gram of tissue for all points inside and outside of any radioactive sphere, for all radioisotopes given in Table 2.

Table 3

Isotope	*A*	*Half-life T in days*	*k*
C	11	0.014	370
N	13	0.007	740
Na	24	0.61	2.8
Cl	38	0.026	163
K	42	0.515	32
V	48	16	0.12
Mn	52	6.5	0.25
Mn	56	0.108	32
Fe	59	47	0.1
Co	56	85	0.03
Cu	61	0.142	46
Cu	64	0.53	50
Zn	63	0.027	170
As	76	1.12	13
Br	82	1.5	1.4
I	130	0.525	4.7
I	131	8.0	1.5
Au	198	2.7	5
Radon	222	3.825	1.0

For those isotopes listed in Table 3, the gamma-ray dosage (including absorption) at the *center* of a sphere of radius R can be taken from Fig. 4 without any calculations. These values are correct within $\pm 5\%$, the variation of the absorption term for the different isotopes. As in the previous tables, A denotes the atomic weight of the isotope; T the half-life of the isotopes is given in days. In the column headed k are listed the number of microcuries destroyed per gram of tissue of each isotope which gives the same number of roentgens at the center of the sphere as 1 μc of radon destroyed/gm of tissue.

The k-values allow direct comparison of the relative gamma-ray intensities per μcd for the different isotopes. This may be of value in consideration of various radiation problems.

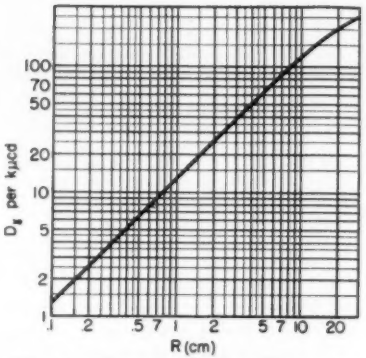

FIG. 4. Gamma-ray dosage D_γ at the center of a sphere as a function of its radius R. The D_γ values correspond to k initial microcuries of any isotope given in Table 3

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THE ORIGIN OF COSMIC RAYS

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THE ORIGIN of cosmic rays has been a mystery ever since their discovery. This discovery was itself a long and difficult process starting, as we look back now, with a paper on residual ionization by Rutherford and Cooke* published in 1903. The understanding that a phenomenon originating outside the earth was being observed did not come until much later—around 1913—when observations of ionization in the upper atmosphere were compared with similar observations at the earth's surface by V. Hess.† At this time it appeared that some form of radiation was responsible for the effects and the adjective "cosmic" came into general use in this connection. Before Hess' work, scientists supposed that the radiation responsible for the effects now known to arise from cosmic rays came from radioactive materials in the earth and in the lower atmosphere. This concept may be called the first theory of the origin of cosmic rays. Many diverse theories have been suggested since that time.

One theory, expressed by the slogan, "birth cries of the atom," achieved considerable popularity with the press. However, as the high energy per particle began to be more thoroughly appreciated, scientists realized that no possible atomic nucleus contained a sufficient mass defect to support this theory. A similar theory requires that the heaviest known atom must so explode as to concentrate its entire energy on a single proton. No stretch of our understanding of physical processes, especially conservation of momentum, leads us to a confirmation of this possibility.

Other theories, presented from time to time, have suggested the occurrence in space of field configurations that are the counterpart of the machines used in earthly laboratories to accelerate particles to high energies. The theory herein presented falls into this category. We shall attempt to account for the existence of cosmic rays and to correlate them with other diverse phenomena, without running into obvious difficulties which have beset previous attempts.

Briefly, we propose a new and tentative theory for the origin of cosmic rays. Cosmic rays consist of the ionized particles that are known to exist throughout space and have been accelerated by transient electric fields in that space. We identify these electric fields as those associated with solar radiation of extremely low frequency. These long-wave radio waves presumably may arise from mechanical turbulence in the highly ionized atmosphere of the sun, in combination with the local and general solar magnetic fields. This idea is attractive because it provides a simple and familiar mechanism for concentrating the energies of many particles of the solar atmosphere upon a single particle in interplanetary space.

This process comes about in a way that does not require thermal equilibrium with the temperature corresponding to the high energy of cosmic

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* E. Rutherford, H. L. Cooke, *Phys. Rev.* **19**, 183 (1903).

† V. Hess, *Akad. Wiss. Wien* **120**, 1575; **122**, 1053 (1913).

rays. We here postulate that the sun is continually radiating a sort of thermal-noise spectrum, whose Fourier-analysis components range in wavelength from approximately 200 miles to 2,000,000 miles. If intensities of the order of one volt per centimeter occur frequently in the neighborhood of the earth, then interplanetary ions can be accelerated to energies as high as 10^{11} electron volts during a half cycle of this low-frequency radiation. Of course, such particles which do not intersect the earth or some other body at the end of a half cycle of radiation will be decelerated approximately to rest in the succeeding half cycle of radiation. Thus we are suggesting that the earth rushes through a tenuous jitter of accelerated and decelerated ions, which we identify as the primary components of cosmic radiation.

We must point out certain apparent difficulties that arise in connection with this theory. To some of them, we can indicate a partial answer. At first sight, one might be inclined to expect that the particles would arrive mainly from the direction at right angles to the vector between the earth and the sun. However, any particle that strikes the earth will have started its acceleration not more than 1,000,000 miles from the earth. Much of its acceleration will have taken place in appreciable terrestrial magnetic fields. Thus, the observed random distribution of cosmic rays merely means that the original particles have a random distribution in space and that the direction of the accelerating electric field is thoroughly masked by the forces of the terrestrial (and possibly solar) magnetic fields. Also, we point out that most of the ions reaching the earth should be positive since the negative charges (electrons) will follow paths of relatively high curvature near the origin of their acceleration, and hence will not so readily achieve the high energies attained by the more massive ions.

This theory avoids the necessity for maintenance of steady electric fields. No individual particles, with energies in excess of 10^{11} electron volts have been observed in connection with cosmic rays.* However, the existence of particles with energies as high as 10^{17} electron volts has previously seemed necessary to account for the total energy of an Auger shower.† These energies are truly enormous. If a baseball were to absorb all the energy from a single cosmic ray of this variety, the ball would roll rapidly across the floor. We find it unnecessary to assume the existence of such energetic primary particles. We account for an Auger shower as arising from a fortuitous fluctuation of the density of ions in space so that a number of ions happens to come down the same configuration of electric and magnetic fields at about the same time. This explanation, if correct, suggests the possible existence of Auger showers of even wider extent than any yet observed.

It seems desirable in addition to point out that a single proton having 10^{17} electron volts of energy is extremely unlikely in the neighborhood of our solar system. Because of the relativistic Doppler effect such a particle would find the visual radiation from the sun seeming very hard indeed. Using the equation for relativistic Doppler effect‡

$$f^* = f \frac{1 - \cos\left(\alpha \cdot \frac{v}{c}\right)}{\sqrt{1 - \frac{v^2}{c^2}}}$$

where f is the frequency of the light in

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* P. Auger, "What Are Cosmic Rays?," p. 60, 88 (University of Chicago Press, 1945).

† Ibid., p. 95, 96.

‡ P. G. Bergmann, "An Introduction to the Theory of Relativity," (Prentice-Hall, New York, 1942).

the frame of reference in the sun; f^* is the frequency of the light in the frame of reference of a particle moving with a velocity v; c is the velocity of light; and α is the angle between the direction of the light ray and the direction of motion of the particle. The factor $\frac{1}{\sqrt{1-\frac{v^2}{c^2}}}$ is the ratio between the apparent mass of the particle in the solar reference frame and its rest mass. A proton having 10^{17} electron volts energy and a rest mass of 10^9 electron volts will have a mass ratio of 10^8. This is the principle factor which multiplies the frequency and the quantum energy of the sun light. Therefore, in terms of solar quanta of about 10 electron volts energy (1000 A.U.), a proton of 10^{17} volts energy is passing through an intense field of radiation having about 1,000,000,000 volts per quantum. This quantum energy is far in excess of the expected threshold for splitting a proton into mesons which soon decay into components. If particles of 10^{17} electron volt energy exist in the neighborhood of the sun, they will soon be broken up into debris by the solar radiation and none of them should live long enough to produce an Auger shower.

The average density of matter in interstellar space amounts to the equivalent of about one proton per cubic centimeter. We have no definite information concerning densities in the neighborhood of the sun. Because of the concentration of meteoric matter, fragments of ejected solar material, tenuous extremities of the solar corona, etc., we should expect the density to be somewhat higher in the solar neighborhood.

The customary formula for the refractive index of an electron gas is

$$\mu = \sqrt{1 - 81N/f^2},$$

for a radio wave of f kilocycles. If we set the electron density, N, equal to one per cm^3, the lower limit, we find that the index of refraction becomes imaginary for all frequencies less than 9 megacycles. It would thus appear, from this simple analysis, that the much lower frequencies herein discussed could not reach the earth.

However, the factor 81, appearing in the above formula, actually represents the quantity:

$$81 = \frac{\epsilon^2}{4\pi^2 m} \times 10^{-6},$$

wherein m and ϵ are the electronic mass and charge, respectively.

The foregoing equations, however, were derived without consideration of the relativity effect. The mass of the electron depends on the velocity. An electron that has attained an energy in excess of 10^6 volts will show a rapidly increasing effective mass, and corresponding diminution of the critical frequency for the transmitted radio wave. We believe that, with electron densities and amplitudes of radio waves which are not unreasonable, we can account for the transmission, through the intervening space, of radio waves originating in the sun.

The theory herein presented is far from proved. There are, however, two reasonable excuses for presenting it at this time. One is that, as will be shown further on, the theory is susceptible to a certain amount of experimental investigation. The other is that the concept may be related to other well-known solar-terrestrial phenomena. For example, part of the ionization of the E and F layers of the ionosphere may arise from the action of the postulated radio waves. The mean free path at these levels will permit the electrons to attain from 10 to 100 volts between successive collisions in the electric field. The same explanation may account for a large part of the light of the night sky and

general auroral glow that goes on more or less continually. We do not intend to imply that the waves cause the large auroral displays, particularly those connected with magnetic storms. It is probable that these latter disturbances are from an entirely different source.

If we extrapolate the effect of low-frequency radiations back to the sun as their source, we come to still more interesting correlations. A field strength of one volt per centimeter at the earth presupposes a field strength in the solar atmosphere such that ions there receive approximately 1,000 volts per mean free path. This condition leads to the existence of an atmosphere of ions and electrons whose apparent temperature is approximately 2,000,000° C. A temperature of this order of magnitude is necessary for interpretation of the spectrum of the solar corona, which, as Edlén has shown, contains lines of highly ionized (10–14 times) iron, nickel, and calcium.

Electrons of this high energy will account for a curious effect that has only recently been observed. Such electrons in the presence of sun-spot magnetic fields will move in circular or spiral orbits with a rotation frequency that depends on the strength of the magnetic field. Zeeman-effect pictures show that sunspots may have magnetic fields with strengths as high as 4,000 gauss. The rotation frequency of an electron in a field of this intensity is such that it will radiate electromagnetic waves having a wavelength of approximately 2½ centimeters. Since fields exist from this maximum down to zero, according to the position with respect to a sunspot, electronic radiation from 2½ centimeters wavelength to a wavelength of hundreds of centimeters should be expected with an equivalent blackbody temperature of approximately 2,000,000° C. Observations at 3 centimeters wavelength, at 10 centimeters wavelength, at 60 centimeters wavelength, and at 300 centimeters wavelength show this type of radiation, whereas at 1.25 centimeters wavelength the equivalent radiation temperature of the sun seems to be about 6,000° C.* It is not known what happens to these radiations during a sunspot minimum, since the apparatus by which these observations can be made has largely been developed during the present maximum.

Of course, it is desirable to give some explanation of how the long-wave radiation which is the basis of this theory can be produced in the sun. Coronagraph movies of the sun graphically portray the enormous turbulence in the solar atmosphere. Since this atmosphere consists primarily of ionized matter, its movements in connection with any magnetic field will produce changes in that magnetic field. If the changes are rapid enough and sufficiently extensive, the radiation will result automatically. The longest wavelength mentioned here, approximately 2,000,000 miles, is chosen because it is the greatest that the sun, acting as a dipole, can radiate efficiently. The magnitude of the general solar magnetic field still requires experimental confirmation. Values of from 30 to 50 gauss are plausible. A change of one percent in this field, at a rate rapid enough for efficient radiation, would produce a field strength at the earth of one volt per centimeter. The field of a large sunspot (4,000 gauss) requires a current of about 10^{13} amperes for its production. One percent fluctuations in this current, at a frequency whose period is comparable to the time required for electromagnetic radiation to cross the sunspot, will cause efficient radiation and produce a field strength of about one volt per centimeter at the earth. We assume

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* G. Reber, Solar Intensity at 480 Mc, *Proceedings of the I.R.E.* **36**, 88 (1948).

that this figure represents a kind of peak value. It is interesting to note that the total radiation expected from the sun involves the radiation of about as much energy as the sun fails to radiate because of the darkness of sunspots.

The foregoing considerations have led to some preliminary experiments with electrostatically shielded loop antennas which indicate the existence of radiation at the earth's surface of about the correct intensity in the frequency range from 25 to 1,000 cycles. Lower frequencies are yet to be investigated. It is not yet possible to say definitely that these experiments show that this radiation comes from the sun. Further experiments are projected to determine this point and the results will undoubtedly shed further light on the theory outlined above.

NUCLEONIC EVENTS

FIRST LABORATORY-CREATED MESONS PRODUCED WITH 4000-TON CYCLOTRON AT UNIVERSITY OF CALIFORNIA

Mesons, nuclear particles heretofore found only in cosmic rays, have been artificially produced for the first time at the University of California Radiation Laboratory. Announcement of this was made last month by Ernest O. Lawrence, director of the Radiation Laboratory, and James B. Fisk, director of research for the Atomic Energy Commission, sponsor of the research.

The liberation in the laboratory of these cosmic ray particles, one of the major objectives of nuclear physicists for the past decade, is regarded by scientists as the most significant event in fundamental nuclear studies since the discovery of uranium fission. Indeed, it may be recorded as one of the great milestones of all time in fundamental nuclear science.

Tracks of the mesons have been observed on photographic emulsion plates placed adjacent to targets of carbon, beryllium or other materials which were bombarded by 380-Mev alpha particles. Only after exhaustive analyses of the tracks was the evidence considered conclusive enough for the announcement to be made.

Professor Lawrence said the development opens up unprecedented opportunities for understanding sub-nuclear processes, particularly the nature of the force which holds the nucleus together and the interaction of nuclear forces. The meson is the best tool ever obtained for examining these forces; its use for the first time under controlled laboratory conditions will enable scientists to study, more or less on their own terms, the critical nuclear processes in which the meson is involved.

But of even greater importance, Dr. Lawrence said, are the portents for the future. He explained that, because the 4000-ton cyclotron possesses just barely enough power to produce low-energy mesons, much larger machines will have to be built to extend the range of investigation.

The actual experiments were performed by Eugene Gardner, research physicist in the Radiation Laboratory, and C. M. G. Lattes, a Brazilian scientist from the University of São Paulo, who came to Berkeley recently on a Rockefeller Foundation Fellowship. (Coincidentally, the Rockefeller Foun-

dation gave the original grant for construction of the cyclotron.)

Dr. Gardner, who received his doctorate at Berkeley and worked on the Manhattan Project at Oak Ridge, has been bombarding photographic emulsions in the California cyclotron for over a year in studies of various atomic disintegrations. In this time he developed methods of working with emulsions in the machine.

Dr. Lattes for the past two years has been working with a group of scientists at the University of Bristol, Bristol, England. Dr. Lattes, Dr. C. F. Powell, and Dr. G. P. S. Occhialini, working at Bristol, have led in the application of specialized techniques for studying cosmic rays by means of photographic emulsions. Drs. Gardner and Lattes and Professor Robert R. Serber, nuclear physicist in charge of the theoretical work in the Radiation Laboratory, explained how the mesons were observed and the part played in this event by the Bristol discoveries.

Dr. C. M. G. Lattes, left, Brazilian scientist, records data as Dr. Eugene Gardner, right, uses a microscope to examine photographic emulsions for mesons. Experiments by the two scientists in the University of California Radiation Laboratory resulted in artificial means

They said that before the Bristol findings were revealed, it was generally believed that only one kind of meson existed. This particle was suggested in 1937 by a Japanese scientist, Yukawa, to explain a gap in the atomic theory of that day; such a particle was later found in cloud chamber experiments by Dr. Carl Anderson, of California Institute of Technology, and he called it a mesotron. The names meson and mesotron have been used interchangeably.

Anderson's meson was found to have a mass of about 200 times that of the electron and either a positive or negative charge of electricity. Found at sea level, this meson has a lifetime of two millionths of a second and energies up to billions of electron volts. It is a secondary cosmic ray particle, and scientists considered that it was made as a result of the bombardment by heavy, energetic primary cosmic rays entering the earth's atmosphere from outer space and colliding with nuclei of the atmosphere.

Drs. Gardner, Lattes and Serber explained that scientists have tried to tailor their theories to fit the idea that only this one kind of meson existed. One of their great difficulties was that this meson did not interact with atomic nuclei; its birth could not be explained adequately, and its brief existence was climaxed by oblivion. But it was obvious that these light mesons were the product of nuclear particle collisions.

During years of careful study and development of special techniques, the scientists at Bristol, taking their emulsions to mountain tops in the Andes and Pyrenees, were able to explain how the light meson often originated from a heavier meson.

The Bristol scientists found on their photographic emulsions, first, mesons of a mass of about 320 times that of

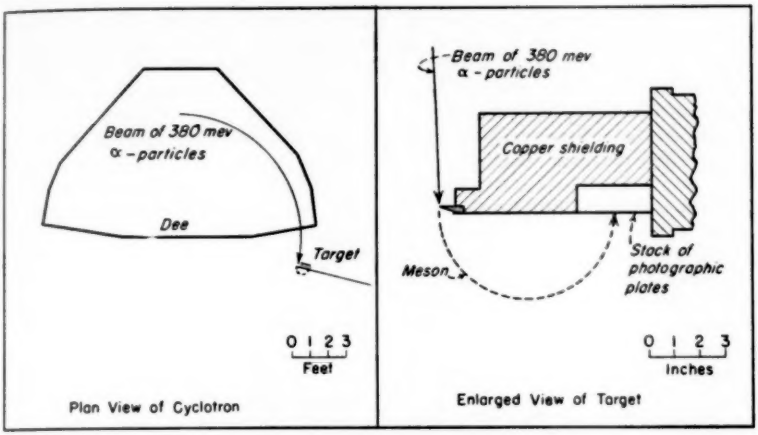


Diagram shows the experimental arrangement which was used in the creation of artificial mesons. At the left, the position of the target and emulsion plate in the cyclotron chamber are shown. At the right is shown an enlarged view of the target and emulsion arrangement. When the target is bombarded by projectiles, mesons are emitted. The magnetic field of the cyclotron deflects the mesons out, and they strike the emulsion placed in the niche at the right. On three sides of the niche are strips of copper, which is an efficient absorber of neutrons. The copper eliminates most of the radiation present in the bombardments, radiation which would obscure the mesons

the electron. They found that these heavy mesons were both positively and negatively charged, and there was some evidence that neutral mesons also existed.

All of these heavy mesons were studied at very low energies, of a few million electron volts, when they were close to or undergoing disintegration. At these energies, the positively charged heavy mesons, being unable to penetrate the electrical barriers of positively charged nuclei, simply wandered through the photographic emulsion until they disintegrated, giving birth to light mesons.

However, the negatively charged heavy mesons were absorbed by nuclei, resulting in the detonation of the capturing nuclei into showers of particles called "stars." At the end of their course, when nearing capture, the mesons made a wavy track. The wavy track is made because the particle is relatively light, and, at the extremely low energies involved, it takes a severe buffeting from nuclei in the emulsion.

Thus the Bristol experiments explained where the light mesons originated and why they were unable to interact with nuclei.

The negative heavy mesons, now known as a result of the Berkeley research to have a mass of 313, are the type which have been produced in the Berkeley cyclotron. The characteristic wavy track and the stars resulting from the detonation of nuclei are also observed. About half the meson tracks observed end in stars.

The mesons were made by placing targets of carbon, beryllium, copper or uranium in front of the alpha particle beam. The negative heavy mesons emitted as a result of nuclear collisions are deflected out by the cyclotron's magnetic field, in a semi-circle away

from the regular path of the positively charged alpha particles in the field.

With protective strips of copper on three sides to keep neutrons and other particles from entering the emulsion, the mesons are nicely separated from other radiations which would otherwise tend to obscure the newly observed phenomena. This experimental arrangement was devised by Dr. Edwin M. McMillan, professor of physics.

Drs. Gardner, Lattes and Serber explained that mesons are actually produced by an intricate sub-nuclear process, which can occur at the peak energy of the cyclotron only a small percentage of the time in a given number of collisions. The explanation is something as follows:

First, an alpha particle is composed of four particles—two neutrons and two protons. On the average, each of these four particles has one-fourth of the 380 Mev which the cyclotron gives to the alpha particle. This equal division of energy is just an average over a given period of time. Sometimes, therefore, when an alpha particle crashes into its target, one of the protons or neutrons has a great deal more than its average one-fourth share of the total 380 Mev. A small percentage of the time one particle will have as much as 225 Mev.

Now, the particles in target nuclei are actually always in motion inside these nuclei. If a proton in a target nucleus happens to be moving toward a bombarding, 225-Mev proton from the alpha particle at the time of a collision, there is a compounding of energy, the total of which is about 390 Mev, just sufficient for the release of mesons.

When two such protons meet head-on, one of the protons is converted, by nuclear exchange forces operating at the time of collision, into a neutron and a negative meson.

The observed mesons have an energy of only about 4 Mev, an extremely low energy for a meson. The mesons travel about 9 inches in a vacuum between their origin in the target and their destination in the photographic emulsion.

The emulsions are about 0.002 in. thick. They are very much like those of ordinary fine-grained photographic plates but considerably thicker. Stacks of about six emulsion plates are placed edgewise to the path of the mesons for ten minutes. The mesons make a track about 0.04 in. long, the track being made by electrons knocked off nuclei in the emulsions. Scientists place the developed emulsions under a 600-power microscope in order to bring the tracks into view.

Dr. Serber said that theoretical calculations have indicated that the mesons were being made from the beginning of operation of the big cyclotron over a year ago. However, since barely enough energy was available and the percentage of meson-producing hits was so small, it was considered a strong possibility that mesons might not be observed at all until an even more powerful machine was built.

However, the compounding of the techniques developed by Drs. Gardner and Lattes permitted the observation of meson tracks on the first plates exposed in the cyclotron bombardments.

Dr. Lattes said that, in British experiments with nature as a laboratory, eight people work a year to get 100 negative meson tracks. At the University of California, one person has observed 27 tracks in 10 minutes of observation.

The first plate exposed by Drs. Lattes and Gardner, for a period of thirty seconds, yielded 100 times as many mesons per plate as were obtained in the Andes in 45 days. This is ten million times as many mesons per second in the cyclotron as on a mountain top.

Improvements have been made in

the technique during the few days since the first mesons were obtained. On the first night, one track in 10,000 was a meson; with improvements, one track in 10 is a meson.

The scientists have placed emulsions inside the cyclotron, where they believe positive heavy mesons, decaying into light mesons, are being made; but so far none has been observed, primarily because of the heavy radiation inside the chamber which obscures the elusive tracks of mesons on emulsions. With the development of new methods, the positive mesons are expected to be found. A positive meson would be created from the collision of two neutrons at these high energies.

Dr. Serber cited the following significant implications in connection with meson production:

1. It proves the idea is right that there is an intimate connection between nuclear particles and mesons and between nuclear forces and mesons.

2. With the means now provided to create and observe mesons under controlled conditions in the laboratory, it may be possible to determine what are the ultimate particles of matter, why only these particles exist, and what their properties are. Dr. Serber said, for example, that it was a simple matter to determine the mass of the mesons, which was of some uncertainty. The mass can be calculated by the radius of its flight in a magnetic field from target to emulsion and the range of its penetration into the emulsion.

3. The study of mesons will be particularly important in determining the nature of the sub-atomic force which holds nuclei together.

4. Techniques will probably be developed, in time, which will enable scientists to study mesons as thoroughly as neutrons and protons. Such techniques would entail the use of specialized Geiger counters, for example.

BRITISH RELEASE REPORT ON PROGRESS AT HARWELL

The Atomic Energy Research Establishment at Harwell, England, has been in existence for approximately two years. On September 20, 1947, *H. W. B. Skinner delivered a lecture to the British Atomic Scientists' Association reviewing the work at Harwell. Below is an excerpt from this lecture which only last month was released to the public* [Atomic Scientists News **1**, 110, (1948)].

The terms of reference of the Research Establishment are to carry out work on research and development in atomic energy. This was interpreted as work primarily aimed towards the development of atomic power and, as a very important secondary objective, towards the provision of radioactive tracers for biological and other research. The task of highest priority was therefore the provision of atomic piles for experimental work and for the manufacture of tracers. Since all aspects of this work have to be covered, from building the pile to questions such as safety of operation, from the manufacture of tracers in the pile to their separation and provision in a form suitable for use, it follows that work has to be done on the following subjects: physics, theoretical physics, chemistry, metallurgy and engineering. Besides these, a division for the control of health had to be set up. For the staffing of the Establishment we had the advantage of the nucleus which was sent out from England into the American project and to the Chalk River Establishment in Canada. This staff, experienced in the work, has had to form the backbone of the Establishment. We were also able to take over blocks of staff from other wartime government laboratories. One of these, from the Telecommunications Research Establishment, Great Malvern, has formed our Electronics Division, and another from the Royal

Aircraft Establishment, Farnborough, has made the nucleus of our Metallurgy Division. Owing to lack of accommodation at Harwell, the first of these two divisions is still working at its original Establishment, though we hope to move to Harwell in the course of the next year. Owing to the nature of the wartime training, especially for radar, we have been able to recruit a very good type of physics staff for Harwell from other government establishments. The physics staff is, therefore, approaching full strength, while staff in the other subjects is still building up as rapidly as it can.

It has been the aim of the Establishment from the start to interpret atomic energy in a broad sense, since it was considered that it would not be possible to build a healthy Establishment if it were confined to a few projects of what might be called nuclear engineering. It was felt that the health of the Establishment would depend on scope being given for fundamental research work. Some major projects undertaken from the beginning fall into this category.

Piles Main Objectives

As has been said, the main objectives of the Establishment were to be the piles, first a pile of very low power for experimental work and control of pile materials and then a pile of higher power for experimental work and the manufacture of tracers. Basic work on these piles had begun before the initiation of the Harwell Establishment, at Chalk River, and in various firms in England. We were thus ready to put the projects in hand and so we were able to announce in August, 1947, that the first pile had started to operate. This pile is known as the Graphite Low Energy Pile or "Gleep." The concrete shield and its accessories had taken about one year to build. About the beginning of July stacking of the graphite began, and it was completed, well ahead of schedule, in about one month. The uranium was then introduced slowly. As the neutron multiplication factor increased with increasing uranium, so the approach of the pile towards criticality could be noted by the readings of a BF_3 chamber recording the flux density of neutrons in the pile. On August 7th, when the pile contained several tons of uranium, the counter recorded 17 neutrons per minute. By August 11th the figure was 55 neutrons per minute, by the morning of August 15th, 2,400 and by the afternoon 6,600. At this point the chamber saturated, the pile being almost divergent. On loading a little more uranium, the pile became divergent and worked at about 0.1 watt, the power increasing exponentially but with a very long time constant. The control rods were then somewhat withdrawn and the time constant increased so that the power rose fairly rapidly to 100 watts, at which point the shut-off rods, which had been set for this figure, operated and shut down the pile. Since the middle of August a considerable amount of work has been done on the operating characteristics of the pile. Recently a simple cooling mechanism has been put into action and the power has been taken up towards the rated figure of 100 kw, and beyond that for short periods. This pile was built for experimental work in physics and on materials, and its flux is small for the manufacture of tracers. However, we shall be able to produce about 5 to 10 curie of radiophosphorus per month, though the low flux will only allow a low specific activity, unless an (n,p) reaction on S^{32} is used, which would reduce the output considerably.

In the next pile to be installed at Harwell (which we hope will work during 1948) the power of operation will be greater (several thousand kw) because the cooling of the uranium will be accomplished by passing a high-speed

stream of air in the channels round the rods. The metal will be sheathed with aluminum to prevent oxidation and to contain the fission products. The emergent gas temperature will be too low for the efficient production of electric power, but it is hoped to make some use of the heat of the pile for warming buildings.

The pile will operate at a neutron-flux of something like 100 times that of the Gleep. This will make possible full production of nearly all the radioactive tracers required for this country. A few useful isotopes will be limited in quantity and a few others only manufacturable by use of a cyclotron. The pile will also provide a powerful source of fission products, distributed among the 35 elements from zinc to gadolinium. At 1,000 kw, about 1 gm per day will be produced. A small quantity of plutonium for experimental work will also be produced.

The pile will also be important in the study of the irradiation of various materials in an intense atmosphere of neutrons. Many of the properties of materials under such circumstances—materials of pile construction, for instance—are not yet known in this country.

Power Piles

These piles will probably be followed by others, more directly concerned with power production. There are many problems yet to be solved. For instance, we must aim at a much more efficient use of uranium-235, the only naturally occurring fissile material, than is contemplated in our existing types of pile. A more fundamental problem is to find out whether we can in some way use uranium-238 and thorium as nuclear fuels. Of course, it is known, for example, that uranium-238 is converted by neutron capture into plutonium, which is certainly a fissile material. But the only known way of doing this is to "burn" uranium-235. However, by the partial replacement of uranium-235 in a pile by plutonium we should be able indirectly to "burn" some part of the uranium-238. In view of the obvious stringency of supplies of uranium-235 (present in natural uranium as only 1 part in 140), it will be important to find out how effectively such a process can be carried out in practice.

Instrumentation

The next group of projects to be considered are those associated with the piles—the measuring instruments for radioactive radiation. These are ionization chambers and Geiger-Müller counters. The ionization chambers are used for pile control and for the general monitoring of radiation in the buildings. The counters are used for the investigation of weaker radioactive radiation. These instruments form an essential part of the equipment, and a great deal of work has been put into them. Much of it has been done at Chalk River. In Harwell, we have set up a group working on counters and ion chambers; in Malvern we have a strong electronics group and we also have a small one in Harwell. The Malvern group has been responsible for the provision of scalers for use with the counters and amplifiers for use with the ion chambers. Many more specialized problems also arise. A number of types of health instruments have been developed. For example one of these is a hand monitor for making sure that the workers who have handled radioactive products have no residual activity left on their hands. Besides this, a number of instruments are being developed which are of great assistance in radiochemical work where a large amount of routine counting is required.

Then, also, there is the radiochemical work associated with the extraction and properties of highly active materials.

At present at Harwell there are no facilities for such work, so we only have an ordinary chemical laboratory, but the necessary chemical laboratories are being built. The ordinary chemical laboratory will be followed very shortly by a "warm" laboratory for moderate activities and next year we shall have a "hot" laboratory for highly active work.

Tracers for Medical Research

One of the main aims of the Establishment is the provision of tracer elements for medical research work, and, as I have said, the piles will supply this need. The counting equipment we are developing will enable the material to be used as efficiently as possible.

It so happened that during the war the development of mass spectrographs was undertaken in this country. These have now become available to us and have led us to branch out into the field of non-radioactive tracers, which are simply enriched specimens of known stable isotopes such as C^{13}. For many biochemical uses these are preferable to the radioactive tracers since they do not involve active doses, and also elaborately prepared organic compounds do not decay away. Further, one element which is important, nitrogen, has no suitable active isotope. The mass spectrograph is the analyzer for work with inactive tracers.

It was therefore decided to put in hand at Harwell work towards the preparation of inactive tracers, the most important of which are D, C^{13}, N^{15}, O^{18} and S^{32}. It was known that N^{15} was being produced in the U.S.A. by a method known as chemical exchange. This is a reaction between ammonia and a solution of ammonium nitrate which tends to concentrate the heavier isotope N^{15} in the solution. The work is done by allowing the gas to interact with the solution, which wets a large area of so-called "packing" in a long vertical column. This process is used commercially in the U.S.A. and we hope that a plant will also be set up in England in the near future. With C^{13} and O^{18} similar processes can probably be used, but there are difficulties. At Harwell we are working on a modified process in which gaseous CO is allowed to interact with liquid CO at roughly liquid air temperature. This process shows considerable promise.

These methods are specialized for particular isotopes, but we have the ambition to have available methods by which the isotope of any element can be prepared in small quantities. The simplest general method is that of thermal diffusion, in which a gas is placed in a long vertical column down which passes a heated wire or rod. The molecules containing the heavier isotope tend to concentrate at the cool outer wall, while those of the lighter isotope concentrate near the hot surface. Thus a convection is set up in which the lighter isotope is concentrated at the top of the tube. Several such tubes placed in series will give the required concentration of 20 to 50 times.

Electromagnetic Separation

Also, we are planning an electromagnetic separation plant which will have the advantage that, in principle, any isotope can be separated even if it is one of many. This is essentially a large, high-current mass spectrograph. A large enrichment can be obtained.

"1947 Summary of Nuclear Data"

Unsettled relations between printers with whom we deal and certain of their employees have forced us to postpone publication of "1947 Summary of Nuclear Data," which was to have been published as part 2 of this issue. The special issue, a dividend to subscribers of NUCLEONICS, will appear when conditions permit.

Thus we hope to separate isotopes of interest to nuclear physics such as K^{40} and Ca^{48} and many others. The quantities readily available will range between fractions of a gram and several grams.

Finally, I must say some words about the plans for pure nuclear physics research. Some lines of work have already been started, but I do not propose to touch on these. I must say a few words, however, about the machines which are constructed or under construction for particle acceleration. These are:

(1) Van de Graaff machine for protons or deuterons of 4 or 5 Mev.
(2) Synchro-cyclotron for protons of 150 Mev energy.
(3) Synchrotron for fast electrons.
(4) Linear accelerator for fast electrons.

Details of Machines

With regard to (1), the machine is already working and a voltage of about 3.75 Mev has been obtained with a pressure of 20 lbs per sq in. of freon and 80 lbs of nitrogen. The problem now is to apply the voltage to the hydrogen ions so as to obtain a beam of fast protons or deuterons for experimental work. When this is done, we shall be able to work on nuclear reactions using protons or deuterons; also from the D-D reaction we have a very convenient source of neutrons of defined energy for experimental work.

With regard to (2), although it is smaller than several machines being erected in the U.S.A., the Harwell cyclotron with 110 in. poles is the largest cyclotron at present under construction in England. We hope that it will be completed in the autumn of 1948.

With regards to the synchrotron, we have no project to build a large machine at Harwell. We have, however, at Malvern a very active group which was the first actually to demonstrate a working synchrotron, thereby producing electrons of 15 Mev from a small betatron designed originally for 4 Mev. A model of a synchrotron for 30 Mev has been designed and will be working shortly; also our group is giving assistance to the Oxford (140 Mev) and Glasgow (300 Mev) synchrotron projects.

With regard to the linear accelerator, the Malvern group has had considerable success. They have had a trial length of accelerator working, which speeds electrons up from 50 kev to 500 kev, and another tube planned for 5 Mev will be going soon. Then the next objective will be the production of a machine for 20 Mev electrons, of a length preferably not exceeding about 6 ft. The linear accelerator has the advantage that the main beam of particles can be got out into the air quite directly.

AEC ANNOUNCES PUBLICATION OF NUCLEAR ENERGY SERIES

The Atomic Energy Commission and Columbia University have announced plans for public distribution and sale of the first two volumes of the Manhattan Project Technical Section of the National Nuclear Energy Series. The volumes will be published by the McGraw-Hill Book Company under a contract with Columbia. A major contributor to the series, Columbia is handling the publication for the AEC and its research contractors. Costs of printing and distribution will be absorbed by the publisher at no expense to the government and distribution will be made through regular trade channels.

The National Nuclear Energy Series will be a compilation of research reports on work started during the war and now carried on as part of the United States atomic energy program. The volumes will cover research which is unclassified—not subject to security

restrictions—or research reports which have been declassified.

Planning of the publication began in May, 1944, and in November, 1945, the project was authorized under the name, "Manhattan Project Technical Series," since the work was done largely under the Manhattan Project, War Department predecessor of the AEC. When it appeared that the series would be a continuing one, the Commission subsequently approved the new name, with those volumes dealing with wartime work designated as the Manhattan Project Technical Section of the National Nuclear Energy Series.

The first two volumes will deal with contributions to medical science resulting from work on the Manhattan Project. The first volume to be published deals with the histopathological effects of radiation, based upon research performed at the University of Chicago Metallurgical Project. The editor for this volume is Dr. William Bloom, a member of the Department of Anatomy at the University of Chicago since 1929, who became chairman of that department in 1941. Dr. Bloom is co-editor with A. A. Maximow of "Textbook of Histology."

The following individuals are listed as authors or co-authors of the various chapters of this volume:

Dorry H. Achacoso, Margaret A. Bloom, William Bloom, P. P. H. DeBruyn, Minnie Heller, Marjorie Ismond, Raymond G. Murray, Mila Pierce, Ruth Pinkney Rhoades, Ray S. Snider, George Svihla, Ella Tyree.

The second volume, on the pharmacology and toxicology of uranium and fluorine compounds, is a report of research conducted at the University of Rochester. Since many compounds and elements utilized in nuclear research were not previously studied extensively, it was necessary to make a complete and detailed survey of their toxic and pharmacological effects.

"ATOMIC ENERGY—1948"

NUCLEONICS readers will be interested in the feature article entitled "Atomic Energy—1948," written by Robert Colborn, McGraw-Hill's distinguished Washington Editor, and appearing in the April 10 issue of *Business Week*.

This is a 6,000-word report in which conflicting views on the progress of the Atomic Energy Commission, and the McMahon Act itself, are analyzed. Mr. Colborn toured all the installations of the AEC and based his report on interviews representing a very broad cross section of opinion.

Reprints of "Atomic Energy—1948" are available at 20 cents. On orders of eleven or more, quantity prices will be quoted on inquiry. Address orders for reprints to Mr. Paul Montgomery, Publisher, *Business Week*, 330 West 42nd Street, New York 18.

Dr. Carl Voegtlin, editor of the toxicology volume, is an authority on pharmacology and toxicology, and served with the U. S. Public Health Service from 1913 to 1940; as chief of the Division of Pharmacology, National Institute of Health; director of Cancer Research; and chief of the National Cancer Institute. He is a past president of the Society of Pharmacology and a member of the Association of Cancer Research and the Academy of Medicine of Washington.

The following individuals are listed as authors or co-authors of the various chapters of this volume:

Thomas B. Barnett, Alexander L. Dounce, Andrew J. Dowdy, H. P. Dygert, John F. Flagg, Frances L. Haven, Harold C. Hodge, Charles Horton, Joe W. Howland, L. S. Kogan, C. W. LaBelle, Tien Ho Lan, S. Laskin, Elliott A. Maynard, Roger G. Metcalf, William F. Newman, James A. Or-

cutt, U. C. Pozzani, Eugene Roberts, J. J. Rothermel, A. Rothstein, C. J. Spiegl, G. F. Sprague, Jr., H. E. Stokinger, D. V. Tiedeman, G. H. Tishkoff, M. J. Wantman, Carrol Weil, Henry J. Wills.

The Manhattan Project Technical Section of the National Nuclear Energy Series will provide a comprehensive summary of the technical development of wartime research and operations climaxed by the dropping of two atomic bombs on Japan. In certain cases research data accumulated up to January 1, 1947, and even later, are included to complete studies only partially finished at the end of the war. Most volumes of the series are written by the experts who made possible the wartime developments in nuclear science, and embody their intimate knowledge of developments on the Manhattan Project. In all, it is expected that approximately sixty volumes will be made available for public distribution over the next two years.

These volumes will cover declassifiable research and development results obtained at all major installations of the Manhattan District, U. S. Army Engineers, and the Atomic Energy Commission, including the Argonne National Laboratory, the Carbide and Carbon Chemicals Corporation, Oak Ridge National Laboratory, Columbia University, Iowa State College, the Kellex Corporation, Los Alamos Technical Laboratory, the Tennessee Eastman Corporation, University of California, the University of Rochester, and many others. Names of staff members from almost every research laboratory dealing with any aspect of science or technology related to atomic energy development appear on the list of authors. During the war these scientists and engineers were assembled at a relatively small number of research centers where they took part in the research which developed the first atomic weapons, and in the process produced a wealth of new knowledge in many fields. That part of this new knowledge which is not withheld for security reasons will become widely available to scientists through the National Nuclear Energy Series.

CONDON ASKS INVESTIGATION BY SENATE-HOUSE COMMITTEE

On March 4th, Dr. Edward U. Condon, director of the National Bureau of Standards, sent the letter below to Senator Bourke B. Hickenlooper, chairman of the Senate-House Committee on Atomic Energy. This letter was written as an appeal for an investigation of the "problem of proper relations of the Government to scientific personnel." It was requested that this investigation be apart from that being conducted by the Committee on Un-American Activities, the committee which directed charges of "disloyalty" against Dr. Condon last month.

The following is the text of the letter:

"As you know, the House Un-American Activities Committee has released a report making insinuations about my loyalty and charges of untrustworthiness as a scientist in the Government service. I am not concerned about these charges as they affect me personally. These same accusations were made in a newspaper story and in magazine articles written by Congressman Thomas (J. Parnell Thomas, Republican, New Jersey) in June of 1947. Before that time and since then I, like other Government scientists, have been investigated and re-investigated by various agencies of the Government and, so far as I know, have been uniformly cleared. In fact, in July, 1947, after Congressman Thomas' articles appeared, I wrote Congressman Thomas and the members of his committee offering to appear before the committee and to help in any way to answer any questions that they might

have concerning me. I received no answer.

"That, however, is not the issue that concerns me. As in the case of many other scientists, Government service is not for me merely a means of makng a livilihood. It is an opportunity to serve my country and its scientific advance.

"What concerns me is the national weakness and disintegration of our scientific position, if this whole question of proper relations between working scientists and their Government is not fully clarified. It seems to me that this should be done at once. Because of the very special position which atomic energy has in matters of scientific research, I therefore respectfully suggest that your committee give this matter a complete hearing and careful study. I have been trying since I came to the Government to develop and improve the staff of our principal Federal agency, the National Bureau of Standards, for fundamental research in physics, chemistry, mathematics and engineering. The work of the bureau in the field of atomic energy is limited, but important, and the vital character of atomic research to our national security and well-being is such that no part of it can be endangered or imperilled for any cause without great damage to the country.

"Frankly, recruiting and keeping scientists in the Government service is not an easy job at best, because of salary limitations, budget difficulties and the notorious amount of administrative detail with which the Federal Government burdens its scientists. But in addition to that there has been a mounting tension of threats, of purges, spy-ring exposures, publicity attacks and sudden dismissals without hearings.

"All of these make scientists increasingly reluctant to work for the Government. They greatly unsettle their minds, and distract them from the creative efforts which we hope to get from them. Because of these conditions it is becoming increasingly difficult to ask a man to accept a position with any scientific agency of the Government.

This is a matter of concern to your committee because such conditions also apply to the operations directed by the Atomic Energy Commission. Only last week a prominent physicist from the laboratories at Oak Ridge came to see me, and told me that a group of them were going to leave and wanted to come to work for the Bureau of Standards. I could give them no assurances that conditions of work in the Bureau of Standards would be any better than the conditions at Oak Ridge as they were described to me. While this person seemed skeptical at the time I am sure he is this week convinced of the truth of what I said.

"An honest reading of the history of the war will show that Germany, Italy and Japan greatly weakened their scientific condition by a series of purges of prominent scientists and a general intimidation and stifling of the spirit of free inquiry among the others. It has been said that after every war the victors adopt the vices of the vanquished. I am sincerely concerned that we are on the point of doing that in the matter of how we deal with scientists in Government.

"The issue is larger than appears on the surface because with the growing inter-relationship of science and goverment there are today very few working scientists (at least in the physical sciences) who are doing work for the Government or associated with institutions deeply involved in this kind of work. Therefore the whole scientific life of the nation is involved.

"I am convinced that your committee, with its established record of careful thorough study of the problems before it, could do the country a great service by studying the whole problem of

proper relations of the Government to scientific personnel. Your committee has justly earned the confidence of the nation's scientists, as well as the general public, by reason of its demonstrated understanding of the nature and importance of the scientists' work. I would hope that an investigation by your committee would result in restoring conditions in which men of intelligence will be willing to work for their government and will not be constantly harassed and harried by irresponsible attacks on their character. It must be remembered that the physicists of this country instituted voluntary security measures in 1939 long before they were able to get their Government to show an interest in atomic energy matters.

"In fairness and justice we must put an end to public smears on scientists in and out of Government, and we must also do it in order not to throttle the scientific research which is so important to the material side of our civilization.

"If your committee should undertake to make this study I assure you that I will be glad to cooperate in every way, and I am confident that scientists and scientific organizations throughout the nation will provide you with every assistance you may desire."

MDDC REPORTS AND ABSTRACTS AVAILABLE FROM AEC

Abstracts and copies of Manhattan District Declassified Documents (MDDC), reports which summarize results of research conducted under the auspices of the Manhattan District and the Atomic Energy Commission, are now available through the Isotopes Division, Oak Ridge Operations, Oak Ridge, Tennessee.

To acquaint interested investigators with the contents of available documents, the Isotopes Division plans to circulate abstracts of them. These abstracts are prepared by the Technical Information Division of the AEC and are published monthly.

Abstracts and reports can be obtained on application to the Isotopes Division. The supply of Volume I is exhausted, but as sections of it are reissued, they will be mailed to all who request them.

44 ISOTOPE SHIPMENTS MADE TO 8 FOREIGN COUNTRIES

Forty-four shipments of radioactive isotopes have been sent abroad to individuals and research groups in eight countries since the start of foreign distribution of radioisotopes, according to an announcement by the Atomic Energy Commission.

The foreign distribution program of radioisotopes was announced on September 3, 1947, by President Truman in a message to the Fourth International Cancer Research Congress in St. Louis. Since that time, 16 countries have completed the necessary arrangements for receiving the radioisotopes.

Countries which have so far made arrangements to obtain the radioisotopes from the uranium chain-reacting pile at Oak Ridge, Tennessee, are Australia, United Kingdom, Canada, Italy, New Zealand, Argentina, The Netherlands, Denmark, Peru, Cuba, France, Belgium, Union of South Africa, Sweden, Ireland, and Switzerland. Of these, shipments have been made to Australia, Argentina, United Kingdom, Denmark, Peru, Canada, Italy and Sweden.

AEC INCREASES SUPPORT OF CANCER RESEARCH WORK

The Atomic Energy Commission is expanding its assistance and support to the nationwide cancer research work by a four-point program which involves radioactive materials, new facilities, and financial aid.

The Commission's program, costing approximately $3,000,000 from funds

authorized by Congress for cancer research, was designed after months of study to explore the possible benefits of atomic energy to cancer sufferers, to assist and augment existing projects, and to avoid duplication of work now being carried on by the U. S. Public Health Service and other government agencies, the American Cancer Society, and a number of privately endowed foundations and medical research centers.

A basic objective of the plan is the development of the use of radioactive materials in studies of the nature of cancer, its diagnosis and treatment. The main points of the Commission's assistance are: (1) provision of radioisotopes for cancer research and treatment by qualified U. S. medical and research workers; (2) support of selected cancer research projects carried on by hospitals and universities in various parts of the nation; (3) establishment of cancer research facilities, including limited hospital space, at four Commission laboratories; and (4) Support of research related to cancer carried on by the Committee on Atomic Casualties of the National Research Council.

BUREAU OF STANDARDS NOTES

The following information on recent research was reported by the National Bureau of Standards:

I-131 in a Comparison Test. Samples of a solution of I^{131} have been distributed to 85 institutions throughout the country for measurement of its specific activity. Reports from about half of these institutions have been received.

The reported values range from 24.3 mrd per ml to 86.6 mrd per ml with one institution reporting a value as low as 1.9 mrd per ml, all values having been corrected to a common date. The value obtained in the Bureau of Standards was 59.8 mrd per ml by comparison with a RaD + E standard.

The necessity for some kind of standard measurement is evident.

Study of X-ray Protection. Some laboratory measurements have been made on the 90 deg scattering from lead, copper and concrete to determine the proportions of the radiation intensity so scattered and its quality, both of these factors being necessary secondary protective barriers. In the region up to 200 kv, it is indicated that the intensity of the 90 deg scattered beam, measured at one meter from the scatterer, is about 0.1% of the incident beam.

Measurements in the high-energy region indicate that this figure will remain about the same up to a million volts, but further work in this connection is under way.

TEXAS CO. MAKES NUCLEAR RESEARCH GRANT TO MIT

A $250,000 grant for atomic research and training of nuclear scientists has been made to the Massachusetts Institute of Technology. The funds will be used for long-range pure research in nuclear fission and related basic studies on the ultimate nature of matter and energy, to construct high-voltage equipment of advanced design, and to train scientists in nuclear theory and its application.

This work will be carried on primarily in the Laboratory for Nuclear Science and Engineering, which will coordinate its efforts with the departments of physics, chemistry, chemical, electrical and mechanical engineering, metallurgy, and biology.

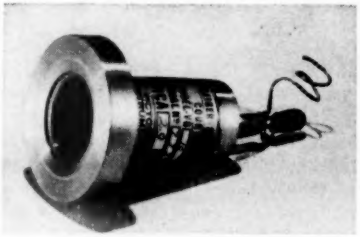
AEC AND MONSANTO CO. RENAME OHIO LABORATORY

The Atomic Energy Commission and Monsanto Chemical Company have announced that the installation to be operated at Miamisburg, O., by the company for the AEC would be officially named the "Mound Laboratory."

PRODUCTS and MATERIALS

G-M COUNTER

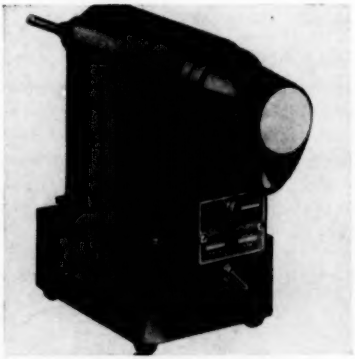
Victoreen Instrument Co., 5806 Hough Ave., Cleveland, O. The VG series of mica window counters have the following specifications: overall length—4¼ in.; flange diameter—2$\frac{7}{32}$ in.; window aperture—1⅛ in.; window thickness (max.)—4.5 mg/cm² (model



15), 3.2 mg/cm² (model 10); plateau—length (min.), 200 v, slope (max.), 5% per 100 v; threshold (approx.)—1200 v; and standard filling mixture—argon with 8% added organic vapor, at approx. 12 cm Hg.

VACUUM MONITOR

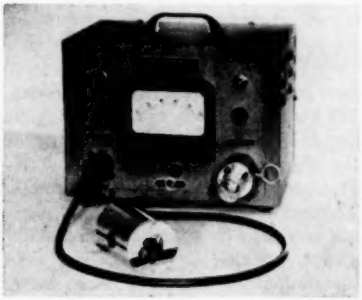
Skaneateles Mfg. Co., 122 Dickerson St., Syracuse 2, N. Y. The type SM Skanascope Vacuum Monitor is an electronic device designed for the accurate indication of vacuum pressures in the 20 to 600 micron range. Its signals are registered by high-visibility luminous tube units, connected in series with a gage tube which, in turn, operates a rated 3-ampere inductive load relay for the actuation of external apparatus such as cutting-in additional pumping capacities, automatic valving, etc. The gage tubes are of the "G" type, their common exhaust connection



being arranged for optional metal-to-glass, rubber-to-glass, or direct glass weld sealing.

MILLIWATTMETER

Rowe Engineering Corp., 2422 N. Pulaski Rd., Chicago 39, Ill. Two methods of measurement are provided in the type MW55, vhf-uhf milliwattmeter. One is a thermistor bridge arrangement for indicating powers from 2 to 60 milliwatts, and the other a PM filament brilliance comparison method for higher powers from 20 to 1000 milliwatts. Power indications



by either method are indicated on a front panel meter and from calibration charts provided. The unit operates from a 105–130 volt, a-c, 60-cycle line and consumes approximately 40 watts. Weight with accessories is approximately 18 pounds.

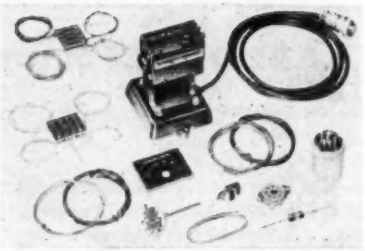
GALVANOMETER

Leeds and Northrup Co., 4934 Stenton Ave., Philadelphia 44, Pa. The type E galvanometer has its moving system, magnet, lamp, scale and lamp transformer in a single, compact case. Available sensitivities are 0.5 microvolt, 0.005 and 0.0005 microampere per mm. The period is about 3 seconds. The instrument has, in effect, an added

low sensitivity. If a deflection shoots the galvanometer's indicating light spot off scale, a secondary spot appears. Moving only one tenth as far as the main spot, this secondary spot shows direction of deflection and approximate magnitude of circuit-balancing adjustments.

CONVERSION KIT

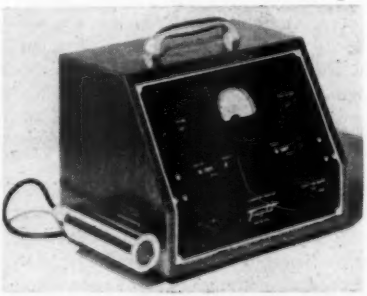
Instrument Development Laboratories, 223 W. Erie St., Chicago 10, Ill. A kit has been designed to convert any of the above company's manually operated scalers into automatic units. This model 3011 kit contains all parts which are to be added to a manually operated scaler, as well as complete instructions for installing these parts.



Installation is said to be simple, since very little circuit change is required. The company also announces that if the scaler is returned to the factory it will be converted for a small additional charge.

CONTAMINATION MONITOR

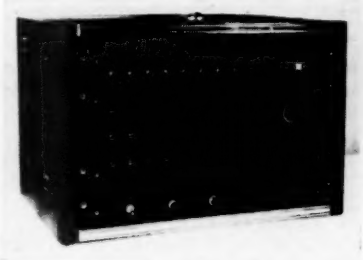
Tracerlab, Inc., 55 Oliver St., Boston 10, Mass. The model SU-3 Laboratory Monitor has been developed as a routine contamination monitor for use in radioactivity laboratories. It is a portable, a-c counting-rate meter of moderate accuracy, with three, full-scale meter ranges of 200, 2000 and 20,000 counts per minute. It comes complete with a thin mica window (3–4 mg/cm^2), sensitive Geiger tube, enclosed in a probe connected to the instrument by a 4 ft length of shielded cable. A clip is provided at the side of the instrument for holding the probe. In addition to a visual indication of counting rate on the meter, a built-in loud speaker provides an aural measure



of the amount of activity which is present.

INTERVAL TIMER

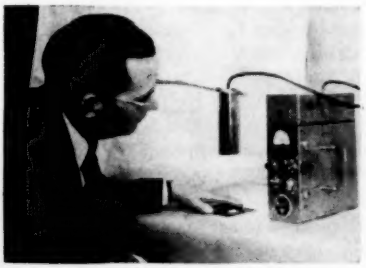
Potter Instrument Co., Inc., 136–56 Roosevelt Ave., Flushing, N. Y. The model 450 Interval Timer has been designed to measure intervals in steps of 0.625 microseconds and to register, directly, intervals up to 1 second. Longer periods can be recorded by use of an external counter to register the number of times the cycle is repeated. The timer is actuated by positive pulses which can be derived from detectors



such as photoelectric equipment and closing contacts. The time base included in the instrument consists of a 1.6 megacycle crystal oscillator. The oscillator, electronic switch and counter decades are made up as individual units which plug into the chassis. Indication is by no means of neon indicator glow lamps.

PROSPECTING TOOL

North American Philips Co., Inc., 100 East 42nd St., New York 17, N. Y. This portable Geiger-counter radiation detector is shown being used in the



laboratory to check the strength of radioactive materials. It can also be carried by means of a shoulder strap when needed in the field for locating radioactive ore deposits and radium which is lost. The Geiger tube is enclosed in a metal cylinder have a retractable section which can be adjusted for detection of hard and soft radiation components.

ABSTRACTS

CHEMICAL PUBLICATIONS

Mass-spectrometer study of the rare gases, V. H. Dibeler, F. L. Mohler, R. M. Reese (National Bureau of Standards, Washington, D. C.), *J. Research Nat. Bur. Standards* **38,** 617–620 (1947). The appearance potentials and isotope abundances of the rare gases were measured and found to be in close agreement with the values obtained from optical measurements.

Enrichment of the heavy silver isotope by ionic migration in α-silver iodide, A. Klemm (Kaiser Wilhelm Inst., Tailfingen), *Z. Naturforsch.* **2A,** 9–16 (1947). Solid AgI was electrolyzed for 20 hours at 230° and the AgI surrounding the anode was analyzed by the mass spectrograph. At the anode, the ratio of Ag^{107} to Ag^{109} was 49:51. The concentration of Ag^{107} increased to 52% at 2.3 cm from the anode.

Physical principles of isotope chemistry, C. F. v. Weizsäcker (Kaiser Wilhelm Inst. Physik, Gottingen), *Angew. Chem.* **A59,** 105–108 (1947). A review.

Detection and measurement of radioactive isotopes in indicator investigations, W. Riegler, *Angew. Chem.* **A59,** 113–118 (1947). A general discussion.

Some anomalies connected with the internal conversion of γ-rays, M. Frilley, J. Surugue, S-T. Tsien, *J. phys. radium* **7,** 350–352 (1946). Calculated values of the coefficient of internal conversion of γ-rays (ratio of the probability of electron emission to that of proton emission when a γ-ray is absorbed) are compared with abnormally high and low values for various elements.

Evidence against spontaneous emission of neutrons by uranium X, H. de Vries, H. Groendijk (Rijks-Universiteit, Groningen) *Nature* **160,** 435–436 (1947). No artificial radioactivity was induced in silver by the radiation of uranium X, showing that the emission of neutrons is practically zero. No anomalous absorption of the γ-rays of uranium X was observed in lead, aluminum, or paraffin if the thin-walled counter-tube was surrounded by a cylinder of lead 0.5 mm thick. The apparent anomaly of the γ-ray absorption previously reported by da Silveira is caused only by the use of a thin-walled counter without a lead shield.

The production of artificial radioactive atoms, W. Bothe (Kaiser Wilhelm Inst., Heidelberg), *Angew. Chem.* **A59,** 108–113 (1947). A review.

Angular correlation effects with annihilation radiation, M. H. L. Pryce, J. C. Ward (Oxford Univ., England), *Nature* **160,** 435 (1947). The coincidence rate as a function of azimuthal angle has been calculated for the two gamma quanta emitted and scattered during the annihilation of a slowly moving, positron-electron pair. The expected coincidence rate is greatest for perpendicular and least for co-planar azimuths. The angle of scatter is plotted against the ratio of coincidence rate for counters in perpendicular planes to those in co-planar planes, and shows that a marked effect should be expected at a scatter angle of 90°. A similar experiment is suggested using photoelectrons ejected by the gamma quanta. A marked correlation of azimuth may be expected in this case too.

Nuclear magnetic resonances at low temperature, B. V. Rollin, J. Hatton, A. H. Cooke, R. J. Benzie (Oxford Univ.), *Nature* **160,** 436–437 (1947). Nuclear magnetic resonance measurements at a frequency of 2 Mc/sec in liquid and solid hydrogen and in alkali halides has been carried out in the liquid-helium temperature region.

Observations on the tracks of slow mesons in photographic emulsions, C. M. Lattes, G. P. S. Occhialini, C. F. Powell (Univ. of Bristol), *Nature* **160,** 453–456 (1947). Examination of plates exposed at a height of 5,500 m yielded forty examples of the production of secondary mesons from charged mesons brought to rest in the emulsion. In some cases, the secondary particle came to rest in the emulsion so that its range could be measured. These measurements provide evidence for the existence of mesons of different mass. It appears that the emission of the secondary meson cannot be regarded as resulting from a spontaneous decay of the primary particle, in which the momentum balance is provided by a photon, or by a particle of small rest-mass. The results are, however, consistent with the view that a neutral particle of approximately the same rest-mass as the μ-meson is emitted.

Nuclear magnetic relaxation, N. Bloembergen, E. M. Purcell, R. V. Pound

(Harvard Univ. Cambridge, Mass.) *Nature* **160,** 475–476 (1947). The observed relaxation times, describing the energy transfer from the nuclear spin system to the whole of the sample, are at least 10^4 times smaller than the values calculated by the Waller theory. A process is described which accounts for the observed relaxation times. The relaxation times were measured by a saturation procedure. The power absorbed by the nuclei relative to the incident power is proportional to the surplus number of nuclei in the state of lower energy. This surplus falls off abruptly if the incident power is increased sufficiently to raise the temperature of the system of nuclear spins.

An asymptotic solution of the fundamental equation of the statistical atom theory, J. I. Horváth (Univ. of Debrecen, Hungary), *Nature* **161,** 26–27 (1948). To avoid numerical methods for the solution of the fundamental equation of Thomas-Fermi's modified statistical method, an asymptotic solution is suggested which yields good approximations when only a few terms are calculated.

Showers of penetrating particles, G. Wataghin (Univ. of São Paulo, Brazil), *Nature* **161,** 91–92 (1948). Showers of penetrating particles were observed by means of eight-fold coincidences between eight counters arranged in four telescopes scattered on an area of 4 m². Examination of the experimental data leads to the conclusion that the majority of mesons are produced in groups (in collisions of primary protons or secondary protons and neutrons with nuclei), with a multiplicity increasing with energy.

Scattering of D-D neutrons by deuterium, J. F. Darby, J. B. Swan (Univ. of Melbourne, Australia), *Nature* **161,** 22–23 (1948). The tracks of 1,500 recoils from D-D neutrons of 2.53 Mev energy were photographed in a cloud chamber filled with heavy methane filled to an expanded pressure of 1.1 atmosphere, and a statistical count of number of tracks versus angle of scattering was made. The distributions to be expected on the assumption of ordinary and exchange forces were calculated. A comparison with the observed distribution shows good agreement with the theoretical curve for ordinary forces.

Physical properties of element 87, actinium K, and its use in the determination of actinium, M. Perey (Inst. of Radium, Paris), *J. chim. phys.* **43,** 269–278 (1946). The disintegration of Act takes place in two ways: Act $\xrightarrow{\alpha}$ Act K $\xrightarrow{\beta}$ Act X and Act $\xrightarrow{\beta}$ Rd — Act $\xrightarrow{\alpha}$ Act X. Of the Act atoms, 1.2% disintegrate with α emission. The β spectrum of Act K is given. By separating and measuring the Act K in equilibrium with Act, the latter may be determined.

Isotopes of greatest abundance, M. A. Levitskaya (Voronezh State Univ.), *Compt. rend acad. sci. U.R.S.S.* **55,** 395–398 (1947). Unstable isotopic nuclei are positive-active or negative-active depending upon whether they contain fewer or more neutrons than the number in the nucleus of greatest abundance. This rule holds in 323 cases, fails in 4, and is not observed in 27.

The influence of the Coulomb field and the electron shell on the form of the radioactive β spectrum, C. Flügge, S. Flügge (Univ. of Göttingen), *Z. Naturforsch.* **2a,** 6–8 (1947). The electron shell has no influence on the form of the radioactive β spectrum for electron energies greater than 50 e-kv. Tables are given for the influences of the Coulomb fields of nuclei.

Efficiency of γ-ray counters, N. Marty (Coll. of France, Paris), *J. phys. radium* **8,** 29–32 (1947). A method for calcu-

lating the efficiency of γ-ray counters is presented, and calculated efficiencies are given for Cu and Al counters for 0.2–5.0 Mev γ-rays. Calculated and experimental values agree within 10%.

A new type of reaction (He^3, He^4), P. I. Lukirskiĭ, M. G. Mescheryakov, T. I. Khrenina (Radium Inst., Acad. Sci. U.S.S.R.), *Compt. rend. acad. sci. U.R.S.S.* **55,** 117–119 (1947). The tracks of accelerated He^3 ions in photographic plates showed sharp breaks which, in two cases out of 100,000, were due not to elastic scattering of light α particles but to He^4 ions.

The meson mass, R. Steinmaurer (Univ. of Innsbruck), *Naturwissenschaften* **33,** 54–55 (1946). Using a cloud chamber at 2300 m elevation, determinations of the meson mass were made. The calculated mass varies from 90 to 230 m_e, indicating that there are two meson masses: approximately 100–120 m_e and approximately 200 m_e.

The absorption and disintegration of mesons at the end of their course, M. Conversi, E. Pancini, O. Piccioni, *Nuovo cimento* **3,** 372–390 (1946). Negative mesons stopped in carbon decay, while those stopped in iron do not disintegrate.

A measurement of the mean path of α-rays from thorium by the photographic method, H. Faraggi (Inst. of Radium, Paris), *J. phys. radium* **7,** 353–356 (1947). Mean path of the α-rays of Th in air at 15° and 760 mm Hg was calculated to be 2.43 ± 0.03 cm.

. . . I. W. RUDERMAN

LIFE SCIENCE PUBLICATIONS

Some biological effects due to nuclear fission, C. A. Tobias, P. P. Weymouth, L. R. Wasserman (Div. of Medical Physics, Univ. of California, Berkeley), G. E. Stapleton (Clinton National Lab., Oak Ridge, Tenn.), *Science* **107,** 115 (1948). Colloidal uranium oxide, containing 95% U^{235}, was injected into mice, which were exposed to a sublethal dose of slow neutrons three days later. All the mice died within three weeks after the bombardment. Control groups, injected with colloidal $U^{238}O_2$ or U^{235}-enriched oxide but not exposed to neutrons, survived. Also surviving was a control group given only neutron radiation and a group injected with the enriched uranium preparation and exposed to X-rays.

The first group of mice developed ruffled fur in 2–3 days after bombardment; in 3–14 days most had arched backs and some developed diarrhea. All lost weight daily. The four control sets of mice lost weight for a few days, but less quickly, and then gained weight, exceeding their original weights. Anatomical, blood, and histological studies are described for all groups. Discussion and calculations are presented, with emphasis on possible applications to cancer therapy.

On the participation of carbon dioxide in the photosynthetic activity of illuminated chloroplast suspensions, A. H. Brown, J. Franck (Dept. of Chemistry, Univ. of Chicago, Ill.) *Arch. Biochem.* **16,** 55 (1948). Previous work has shown that suspensions of tobacco chloroplasts, on being illuminated, release a burst of oxygen and then a small, gradually diminishing amount of oxygen for roughly one hour. The presence of carbon dioxide increases the rate of the second oxygen production. To determine whether this extra production is caused by a small amount of photosynthetic reduction of carbon dioxide, radioactive carbon was administered as $NaC^{14}O_3$. No C^{14} fixation or reduction was found.

Aspects of the biologic decay periods of sodium in normal and diseased man, G. Burch, S. Threefoot, P. Reaser

(Dept. of Medicine, Tulane Med. School and Charity Hospital, New Orleans) *Science* **107,** 91 (1948). The long-lived radioisotope of sodium, Na^{22}, was injected into 12 subjects and the urinary and blood radioactivity levels determined daily for from 30–60 days. Results are given for normal people and subjects with congestive heart failure and chronic glomerular nephritis. Turnover rates were found to be much lower in the two diseased groups. Other influences found were sodium intake, desoxycorticosterone acetate, mercurial diuretics, water intake, and pitressin.

Concurrent use of radioisotopes of calcium and phosphorus in the study of metabolism of calcified tissues, W. D. Armstrong, C. P. Barnum (Dept. of Physiological Chemistry, Univ. of Minnesota, Minneapolis) *J. Biol. Chem.* **172,** 199 (1948). Ca^{45} lactate and P^{32} phosphate were administered together to a rat; the animal was sacrificed after five days. To analyze the different tissues for each radioisotope, it was necessary to develop a method of separating the two elements. Calcium was precipitated as the oxalate and the filtrate used for the P^{32} measurements. The uptake and retention of both isotopes in calcified tissues were found to be (in decreasing order): femur marrow, femur epiphysis, femur diaphysis, incisor tooth dentin, incisor tooth enamel, molar tooth dentin, and molar tooth enamel. The two radioisotopes do not exchange at equal rates in some tissues, possibly because of differing rates of diffusion.

Rate of penetration of electrolytes into nerve-fibers, M. A. Rothenberg, E. A. Feld (Dept. of Neurology and Biochemistry, Coll. of Physicians and Surgeons, New York) *J. Biol. Chem.* **172,** 345 (1948). Giant nerve-fibers of squid were immersed in artificial sea water containing a radioisotope, and the axoplasm then extruded and analyzed for radioactivity. Results with sodium, potassium, and calcium are reported. The radioactivity level in all three cases was found to rise quickly and then taper off. When the potassium concentration outside the nerve was doubled, the rate of penetration doubled. (The use in this paper of the term "concentration" to describe the radioactivity inside the nerve is misleading, since only *total* activity was determined).

The source of urea carbon, C. G. Mackenzie, V. du Vigneaud (Dept. of Biochemistry, Cornell Univ. Med. Coll., New York) *J. Biol. Chem.* **172,** 353 (1948). A rat was fed L-methionine, labeled with C^{14} in the methyl group, and the respiratory carbon dioxide and urinary urea carbon isolated (the latter with urease). Equal specific activities were found in the carbon atoms of both CO_2 and urea in each of the first two days' samples. This indicates that the carbon of urea is derived entirely from body carbon dioxide, which is assumed to be in equilibrium with respiratory carbon dioxide.

Determination of plutonium in human feces, E. Maxwell, R. Fryxell, W. H. Langham (Los Alamos Scientific Lab., N. Mex.) *J. Biol. Chem.* **172,** 185 (1948). To determine the rate of plutonium excretion *via* the feces, a relatively accurate and consistent method of analysis was used: The feces are dried and then ashed in a muffle furnace. The ash is extracted with hot 4 *N* hydrochloric acid, hydrofluoric acid added, and the mixture taken to dryness. The residue is extracted with 4 *N* HCl and the plutonium reduced by heating with hydroxylamine at pH 0.4–0.7. Addition of oxalic acid precipitates calcium oxalate, which carries down the plutonium. This is digested with fuming nitric acid, taken up in 2 *N* HCl and hydroxylamine and iron added. The

cupferride is formed and extracted into chloroform. The complex is destroyed with $HNO_3 + HClO_4$ and the plutonium again reduced. LaF_3 is then precipitated, carrying the plutonium down. The plutonium content is determined by measuring the α activity of the precipitate. Recovery is 80% or better.

Creatine and creatinine metabolism in the normal male adult studied with the aid of isotopic nitrogen, H. D. Hoberman, E. A. H. Sims, J. H. Peters (Dept. of Internal Medicine, Yale Univ. School of Medicine, New Haven) *J. Biol. Chem.* **172,** 45 (1948). An amount of 1.295 gm of N^{15} creatine was divided into 20 parts and each dose ingested every half hour by an adult male on a diet practically free of creatine. Urine was collected for 38 days and the creatinine content determined periodically. Creatinine samples were also isolated for isotopic analysis. (Creatinine N^{15} concentration has been shown to equal that of the body creatine.) From the results obtained it was calculated that about 0.18% of the body weight is creatine and that the time for 50% turnover is 42 days. About 3% of the administered creatine was converted to substances other than creatinine.

On the 39th day, 1.16 gm of N^{15} guanidoacetic acid were ingested and further urine collections made. Beginning 11 days later, 24 gm of non-isotopic creatine were eaten in small portions over a 5-day period. Similar analyses gave rise to data which are interpreted to suggest that exogenous creatine slows the synthesis of endogenous creatine. Only about 70% of the administered guanidoacetic acid was converted to creatine, N^{15} being found in urinary ammonia and urea. These and other results are discussed at length.

Two typographical errors were noted: Eq. 3 should read

$$\frac{dX}{dt} = -\frac{keC}{100}$$

and Eq. 7 should have a minus sign on the left side.

Effects of X-radiation on sodium thymus nucleate, B. Taylor, J. P. Greenstein, A. Hollaender (National Cancer Inst. and Lab. of Industrial Hygiene, National Inst. of Health, Bethesda, Md.) *Arch. Biochem.* **16,** 19 (1948). To understand better the nature of mutation production by X-rays, solutions of the sodium salt of desoxyribonucleic acid, a prominent component of chromosomes, were exposed to X-rays under varying conditions. The high anomalous viscosity found in unirradiated samples is lowered considerably, the drop continuing several hours after irradiation is discontinued. Apparently X-rays cause the long fibrous particles to break into shorter fragments. Originally monodisperse, as measured in the ultracentrifuge, the nucleic acid becomes polydisperse, indicating wide variation in the size of the fragments. The degree of breakdown depends on the number of roentgens administered, and not on the length of the interval during which the dose is delivered.

Egg albumin furnishes temporary protection, while a lowered temperature decreases the rate of viscosity drop after irradiation. Apparently small fragments are not produced since dialysis resulted in no more than 1% loss. Flow birefringence and alcohol precipitability were found to be lost, but enzymatic susceptibility, pH, refractive index, and extinction coefficient were practically unchanged. The depolymerizing effect of X-rays does not seem to be due to formation of hydrogen peroxide in the solvent.

An improved method for the preparation of desoxyribonucleic acid from calf thymus is also given.

. . NORMAN S. RADIN

PHYSICAL PUBLICATIONS

Delayed neutrons from fission of U^{235}, D. J. Hughes, J. Dabbs, A. Cahn, D. Hall (Argonne National Lab., Chicago, Ill.), *Phys. Rev.* **73,** 111–124 (1948). An electronically controlled pneumatic transfer device known as the "rabbit" was used to transfer a sample of uranium from a region of high neutron flux to a position near a well-shielded neutron counter in a time shorter than the periods of delayed neutron activities whose measurement was desired. The cartridges were ejected from the pile and stopped in a block of paraffin which slowed the neutrons. They were then counted by a BF_3 counter. Scaler pulses were recorded on an electrocardiograph tape; light passing through a string galvanometer focuses an image of the string on a slit, past which sensitized paper travels at a constant speed. By varying the distance of the counter from the sample in the paraffin, the energy of the delayed neutrons were obtained (using the slowing-down relation for neutrons in paraffin). These energies are given along with the half-lives and yields relative to the total neutron emission. The application of the delayed neutron data to pile theory is discussed.

The polarizability of the neutron in an electrostatic field, K. M. Watson (Univ. of Iowa, Iowa City, Ia.), *Phys. Rev.* **73,** 125–126 (1948). On the basis of meson theory, a neutron's meson-charge cloud should have a dipole moment in an electrostatic field. This produces a scattering effect on charged particles, which is, however, much smaller than the effect of the extended cloud itself. The interaction is too small to account for the energy level shift of the hydrogen atom.

A determination of the $S^{32}(n,p)P^{32}$ cross section for neutrons having energies of 1.6 to 5.8 Mev, E. D. Klema, A. O. Hanson (Los Alamos Scientific Lab., N. Mex.), *Phys. Rev.* **73,** 106–110 (1948). Monoenergetic neutron fluxes were obtained by different reactions in various energy regions. The uncertainty in energy varied from 2% to 6%. Irradiation of S^{32} with the neutron flux produced the beta-active P^{32}, and measurement of the activity with a counter indicated the relative cross section for the reaction at various neutron energies. To obtain an absolute value for the cross section, the efficiency of the counting geometry used was determined by using a source whose total activity had been determined. A theoretical curve calculated on the basis of the variation of proton penetrability (through the Coulomb barrier) with energy agreed in general with the experimental curve.

The multiple production of mesons, H. W. Lewis, J. R. Oppenheimer, S. A. Wouthuysen (Univ. of California, Berkeley), *Phys. Rev.* **73,** 127–140 (1948). The impacts of nucleons of high energy of motion ($\gg Mc^2$) on other nucleons lead to meson emission, which is multiple (multiplicity increasing with energy) for strong coupling of the meson fields to the nucleon. A more quantitative description is given by setting up an analogy with the Bloch-Nordsieck theory of radiation by collision, and also by using a kind of perturbation method. Total cross sections in these theories approach 10^{-28} cm^2, and an expression is given for the multiplicity as a function of energy.

Radiations from scandium-46 and titanium-51, C. E. Mandeville, M. V. Scherb (Bartol Research Foundation, Franklin Inst., Swarthmore, Pa.), *Phys. Rev.* **73,** 141–145 (1948). The energies of the beta and gamma radiations from these isotopes were measured by absorption. Coincidences between betas and gammas and between gammas and gammas were measured to obtain the disintegration schemes. For Sc^{46} a

beta followed by two gammas is indicated; for Ti^{51} the residual V^{51} nucleus is left in an excited state, and some or all of them emit two or more gamma rays in cascade.

Scattering of high-energy neutrons by protons, M. Camac, H. A. Bethe (Cornell Univ., Ithaca, N. Y.), *Phys. Rev.* **73,** 191–195 (1948). The scattering of neutrons by protons at energies from 20 to 80 Mev was theoretically computed, using the exact formula for the phase shifts. The tensor forces were neglected, and a square well form of the potential assumed, using ordinary forces, exchange forces, and the symmetric forces of meson theory, which are proportional to $\vec{\sigma_1} \cdot \vec{\sigma_2} \vec{\tau_1} \cdot \vec{\tau_2}$ ($\vec{\sigma}$ = spin, $\vec{\tau}$ = isotopic spin). Ranges of 2.8 and 2.0×10^{-13} cm are employed for the nuclear forces. The depth of the potential well was fixed by the deuteron binding energy and the strength of the singlet interaction by the *n-p* cross section at epithermal energies (2.19 Mev and 20 barns, respectively). Comparison with experiments at Berkeley seems to exclude ordinary and favor symmetric forces.

The scattering of neutrons of energy between 12.0 and 13.0 Mev by protons, J. S. Laughlin, P. G. Kruger (Univ. of Illinois, Urbana, Ill.), *Phys. Rev.* **73,** 197–207 (1948). Cloud chamber measurements on the recoil of the protons gave the differential scattering cross section as a function of angle. High-energy neutrons, produced in a deuteron-deuteron reaction, were incident through a collimator on methane gas at a pressure of 25 atmospheres in a cloud chamber (5¾ in. diameter by 3 in. high). A cam system controlled the cycle; illumination was by capillary arc; 60% isopropyl alcohol was used as the vapor. A reprojection system and measuring engine were used to obtain the length, recoil angle, and azimuthal angle of the proton recoil.

Range-energy relations for protons in substances containing C, H, O, A, and Xe, J. O. Hirschfelder, J. L. Magee (Los Alamos Scientific Lab., N. Mex.), *Phys. Rev.* **73,** 207–213 (1948). Using Bethe's method for the calculation, with screening constants and oscillator strengths obtained from various sources, the stopping power of protons in C, H, O, A, and Xe was obtained. Range-energy relations are calculated by numerical integration for protons in glycerol tristearate, paraffin, argon, and xenon.

Momentum conservation in the beta-decay of P^{32} and the angular correlation of neutrinos with electrons, C. W. Sherwin (Univ. of Illinois, Urbana), *Phys. Rev.* **73,** 216–225 (1948). Monomolecular layer sources of P^{32} were used. The time between the detection of an electron and that of the detection of the recoil ion at a certain angle to it was measured. (An electron multiplier was used to detect the recoil nucleus.) The number of recoils as a function of recoil momentum was plotted for various angles between the electron and the recoil, and compared with the results to be expected for various electron-neutrino correlation functions. A cathode-ray tube displayed a horizontal sweep upon detection of an electron, the recoil nucleus causing a vertical deflection when detected. This allowed a wide range of recoil spectrum to be measured at one time.

It was shown that momentum is not conserved between electron and recoil alone, and a $(1 - \beta \cos \theta)$ correlation function is tentatively suggested.

Angular distribution of the (d,p) reactions making two low states of O^{17}, N. P. Heydenburg, D. R. Inglis (Carnegie Inst., Washington, D. C.; Johns Hopkins Univ., Baltimore, Md.) *Phys. Rev.* **73,** 230–240 (1948). Bombardment of thin targets containing oxides by deuterons gave two sets of

protons, a long-range group corresponding to an O^{17} nucleus in the ground state, and a short-range group corresponding to the first excited state. At low bombarding energies, the short-range protons were ten times as numerous, but at high energies they were about equal in number. The angular distribution of the two groups of protons as a function of the bombarding energy was investigated. The protons were detected by an ionization chamber, linear amplifier, and discriminator; the deuteron beam was monitored by integration of the current. Such information may make possible a comparison of the angular momentum properties of the two lowest states of O^{17}, which probably approximates closely a single-particle model.

Angular distribution from $Li^7(p,\alpha)\alpha$, N. P. Heydenburg, C. M. Hudson, D. R. Inglis, W. D. Whitehead (Carnegie Inst., Washington, D. C.), *Phys. Rev.* **73,** 241–245 (1948). The distribution of the recoiling alpha particles in angle is of the form $Y(\theta) = Y(90°)(1 + A\cos^2\theta + B\cos^4\theta)$. A and B are obtained from the data as a function of bombarding energy from 1 to 3 Mev. A decreases with energy from 2.1 to 0.7 and is positive; B is negative and of the order of 0.5, but its energy dependence is uncertain.

The cosmic-ray counting rate of a single Geiger counter from ground level to 161 kilometers altitude, J. A. Van Allen, H. E. Tatel (Applied Physics Lab., Johns Hopkins Univ., Silver Spring, Md.), *Phys. Rev.* **73,** 245–251 (1948). A single Geiger counter was sent aloft in a V-2 rocket at 41° N. geomagnetic latitude in order to determine the counting rate of cosmic rays as a function of altitude. Pulses were radio-telemetered to a ground station. A set of counters behind the single counter gave the effect of multiplication in the rocket when connected in coincidence with the first. This effect was found to be almost independent of altitude. The counting rate increased with altitude until only 58 gm/cm² of atmosphere were above it. It then decreased to an altitude corresponding to 1 gm/cm², where it leveled off. A height of 161 km was attained, and a reasonably flat plateau exists from 50 km to that point. The counting rate observed on the plateau corresponded to a primary flux of about 0.1/sec/cm²/steradian (assuming isotropic charged primary flux).

Analysis of the impulses from Geiger-Mueller tubes, S. C. Curran, E. R. Rae (Univ. of Glasgow, Scotland), *Rev. Sci. Instr.* **18,** 871–876 (1947). An interval-selector circuit was used to determine the dead time and the frequency of spurious counts due to positive ion bombardment. Plateau slope was also investigated. The circuit is particularly useful for tubes containing insulation beads along the central wire. It works by finding the coincidence rates between the Geiger pulses and those of a differentiated square wave, as a function of the duration of the square wave. For durations less than the dead time, there should be no coincidences. Results are given for oxygen, nitrogen, hydrogen, and methane-alcohol counters.

A precision alpha-proportional counter, J. A. Simpson (Argonne National Lab., Chicago, Ill)., *Rev. Sci. Instr.* **18,** 884–893 (1947). To determine with accuracy the number of alpha particles given off by a sample, a proportional counter operating with a constant flow of 99% methane gas was designed. A short resolving time allows the use of the counter in the presence of a very high beta-ray background (up to 5×10^9 soft betas per minute), since there is very small probability that enough betas will be counted during a single resolving period to register as a

large (alpha) pulse. A gas lock allows the insertion of a sample into the counting volume, 50% of the disintegrations from a thin radioactive sample being registered by the counter.

Calibration of a Ra + Be neutron source by the B(n,α)Li reaction, F. G. P. Seidl, S. P. Harris (Argonne National Lab., Chicago, Ill.), *Rev. Sci. Instr.* **18,** 897–899 (1947). The rate of helium production in the above reaction was measured by noting the gamma activation of manganese, after finding the ratio of the gamma activation to the helium production for a known mixture of B and Mn. Knowing the distribution of slow neutrons in the boric acid solution, the probability of neutron capture can be computed, and from it and the He produced, the neutron flux can be determined.

R-F system for frequency modulated cyclotron, K. R. MacKenzie, V. B. Waithman (Radiation Lab., Univ. of California, Berkeley), *Rev. Sci. Instr.* **18,** 900–907 (1947). As a test of the problems to be encountered in the 184-in. cyclotron, a single dee resonant system and grounded grid oscillator were used on a 37-in. cyclotron. A rotating condenser was used to provide the frequency modulation. Deuterons of 7.5 Mev were accelerated by a dee voltage up to 15 kv, the frequency ranging from 9.5 to 12 megacycles; protons of 15 Mev were accelerated by dee voltages up to 11 kv, the frequency ranging from 19 to 24 megacycles over a modulation cycle.

Electronic sequence control circuit for the operation of a Wilson cloud chamber, F. L. Allen (Univ. of Chicago, Ill.), *Rev. Sci. Instr.* **19,** 24–27 (1948). The circuit has the following basic parts: a coincidence circuit to select the event to be photographed, a circuit to operate a valve producing the expansion, a time delay to operate before the illumination, a clearing field circuit, and a blocking circuit to prevent re-expansion before recovery of the chamber. Adjustable time delays are made of unbalanced multivibrators. To operate the expansion valve, two thyratrons are used.

Unidirectional pulse operation of a 22-Mev betatron, H. W. Koch, C. S. Robinson (Univ. of Illinois, Champaign), *Rev. Sci. Instr.* **19,** 36–39 (1948). Particularly for use with a cloud chamber, operating about every thirty seconds, unidirectional pulse operation rather than flux-biased continuous operation is indicated by power considerations. A circuit is described which sends a pulse of current, approximately sinusoidal, through the betatron magnet coil. It is produced by a condenser discharge through a mercury-pool switching tube and the coil.

The measurement of radioactivity in solution, A. K. Solomon, H. D. Estes (Harvard Med. School, Boston, Mass.), *Rev. Sci. Instr.* **19,** 47–50 (1948). A glass tube counter is dipped into a cup containing the radioactive solution in order to determine the activity. The optimum clearance between the counter and the cup (for best combination of total count and count per ml of solution) is determined. Comparison with other methods of determining activity indicates that this is a good method for beta energies above 1 Mev.

Theory of the study of transfers within a multi-compartment system using isotopic tracers, C. W. Sheppard (Vanderbilt Univ., Nashville, Tenn.) *J. Applied Phys.* **19,** 70–76 (1948). A system of n compartments, in which a questionable amount of substance S has moved from one compartment to another, is discussed. The quantity S can be determined in a single experiment from measurements of the amounts of n-tagged species in the individual compartments. Application is made to studies using multiple isotopic tracers.

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